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ANN Modeling and GA Optimization of Zinc Removal from Wash water by Electro-coagulation Process

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***Abstract*—**The present research concentrates on Artificial Neural Network (ANN) for modeling and Genetic Algorithm (GA) for optimization of zinc removal from the industrial wash water by EC process. In the EC process, the most important control independent variables are Initial Concentration (Ic – 216.5 to 866mg/L), Current Density (Cd – 0.1 to 0.6A/dm²) and Time (T – 2 to 15mins). These variables are also affecting the performance of zinc removal. ANN model was able to predict the maximum removal of zinc with two transfer functions like tan sigmoid at hidden layer with 8 neurons, purelin at output layer. Feed forward multilayered perception with Levenberg - Marquardt back propagation training algorithm was used for train the design with Mean Squared Error (MSE) of 1.18 and Correlation Coefficient (R2) of 0.9909 in ANN shows that the model was capable to predict the zinc removal. Single Objective Optimization for maximizing the zinc removal was conducted using GA over the ANN model. Using pattern search method in a GA, the best optimum conditions are recorded as 217.5 mg/L, 0.1A/dm² and 2mins for Ic, Cd and T respectively and the maximum zinc removal at the above condition as 88.71%.

Keywords-*Electro-coagulation process, Artificial Neural Network, Genetic Algorithm, Wash water, Zinc removal.*

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I. INTRODUCTION

All Now a day, electroplating industries are discharging the huge amount of toxic pollutants contained wash water into the river, seas, candle, lake and other natural sources, etc., Result of this, a significant threat to the environment and human health. Excessive level of heavy metal especially zinc present in the wash water may leads the throat dryness, chills, cough and vomiting, etc., Zinc level should be maintained in the water as 5ppm which is standardized by FDA (Reshmi G Saseendran et al, [1]).

Zinc is the one of the most important toxic and dangerous hazardous metallic pollutants in the wash water from the outlet of electro plating industries. There are many treatment methods and techniques on how to remove the zinc from the wash water. Large amount of zinc is presence in the galvanization, paint, pigment and electro plating industries wash water and it is not properly removed from this industrial waste water before discharging to the environment because of this huge problem in human and animal health and also ecology system (Charif Gakwisiri et al, [2]).

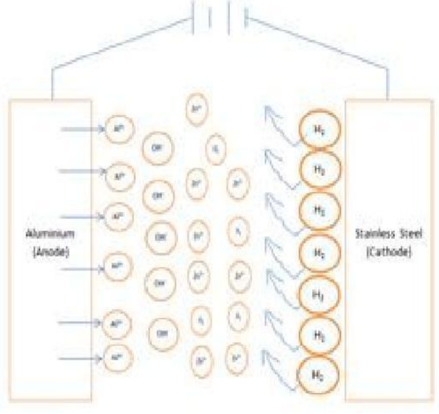

Figure 1 Conceptual sketch of the EC mechanism

Among the various wastewater treatment methods and techniques, electro coagulation is the most effective, quick and promising technology for treating the plating industrial wash water. The conceptual mechanism which is employed in the electro coagulation process is shown in the below figure1 (J.A.G.Gomes et al, [3]).

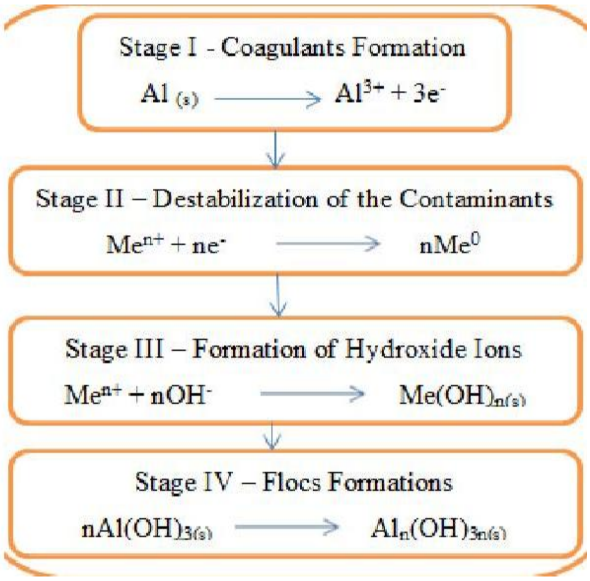

Figure 2 Stages of reactions occurring in the process

The figure 2 shows the three stages of ion producing pattern happening in the electro coagulation process and the reaction equations (I. Heidmann et al, [4]). Nafaa Adhoum et al. [5] conducted a study on the removal of zinc from the electro plating industries by electro coagulation and concluded that electro coagulation treatment technique/method could be the most effective and best method/technique used to remove the heavy metal in general and particularly zinc from the wash water of electro plating industries. Ahmed Samir Naje et al.

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[6] have described in details of various methods and applications of electro coagulation technology in both domestic and industrial wash water treatment and also gave a noteworthy outlook of heavy metals removal modeling methodologies and the investigation about the optimization of process parameters with the economic considerations. As per the research people statements, the electro coagulation method has the ability to develop the model by using block oriented approach and the process has a nonlinear model structure, being well suited to be used as models in system identification (A.Genc et al [7]).

Last two decades, the advent of ANN which has been effectively applied to countless fields of engineering, medical sciences, economics, meteorology, psychology, neurology, mathematics and others (S.L.Pandharipande et al [11]). Owing to their uncomplicatedness towards simulation, prediction and modeling (Ammar salman dawood et al [19]). ANNs have been developed as an overview of mathematical models of neural biology (Shilpi Rani et al [14]). For practical approach, ANN was applied for learning different functions such as functions with real discrete and vector values (Saeid Pakrou et al [15]) and for process modeling., Application of ANN has been successfully engaged in environmental engineering, because of reliable, robust and salient features in taking the nonlinear relationships of parameters in complex systems (Abhishek kardam et al [16]).

In chemistry, the relationships between variables are almost very complicated and highly nonlinear. One of the most reliable methods to be ANN. In fact, ANN is very powerful in dealing with nonlinear type. Most of the publications have underlined the interest of using the ANN (Mohamed NOHAIR et al [17]). During the electrochemical process, removing pollutant from wash waters mechanism is very complex and not in clear idea. If the process mechanisms are not clear, for the modeling purpose, the conventional methods are not also suitable. For classical process modeling, ANN as the best alternative tool because they do not need detailed information systems and while trained, needed only the input-output data sets, to provide good result in the ANN link pattern approach (Ciprian G. Piuleac et al [18]). Numerous reports are available with respect to ANN, applications for wide range of problems in water and wash water treatment process. Recently researchers have successfully modeled a three layer feed forward BP neural network to predict the removal of Cu(II) from industrial leachate by pumice and Zn(II) from hazelnut shell. Z (D.Gnanasangeetha et al [20]).

GA-based optimization is a stochastic search method that involves random generation of positional design solutions which it systematically evaluates and refines until a stopping criterion is met. Traditional optimization methods are not suitable to solve problems where the formulated objective functions and constraints are very complicated and implicit functions of the decision variables. Compare with conventional optimization techniques, GA is a robust, global and can be applied to domain-specific heuristics (Rupen Phipon et al [28]). GA can be used in general, indifferent and unconventional optimization problems. For machine learning - function optimizes and system modeling, GA is the best optimization technique (Doriana M.D Addona et al [29]). Cao and Yang et al., carried out an experiment, and then they have used artificial neural network (ANN) and genetic algorithm (GA) together to establish the parameter optimization model.

In the present research paper deals with the applicability of artificial neural network for modeling and genetic algorithm for optimization on removal of zinc in the industrial wash water. Based on the experimental study, we recommended three layers (input, hidden and output). ANN model with different transfer functions using a back propagation feed forward and LM algorithms to predict the removal of zinc in percentage from the industrial wash water by electro coagulation process with GA for developing an excellent optimization procedure. First, we studied the relationship between the input parameters and output response by ANN modeling technique. Secondly, the optimum condition of three input parameters (initial concentration, current density and time) and output response were identified. Last but not least, to compare the outputs obtained from the model with the experimental and the methodology of the process.

II. Experimental Methods

Composition of Barrel Plating Wash Water (Zinc Sulphate Bath: Zinc Sulphate (0.20M), Sulphuric acid (0.01M), Sodium Sulphate (0.4M), Boric acid (0.16M), Anode (graphite) & pH (2.75). The following Instruments are used for the experimental setup (figure 3), D.C. Power supply with ammeter and voltmeter, magnetic stirrer, spectra lab (speed), aluminum (anode), mild steel (anode) & stainless steel (cathode).

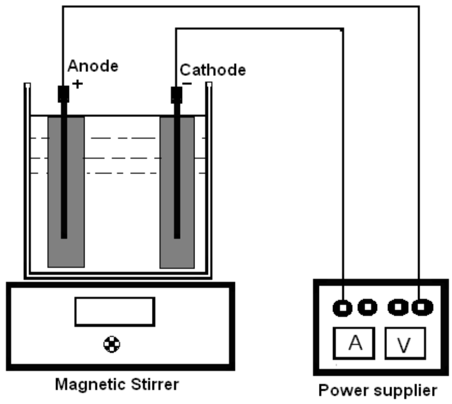

Figure 3 Experimental Setup

The electrode used in this study consists of aluminum plates and stainless steel plates of 99.99% purity. All the chemicals were of analytical grade and the reagents were prepared using double distilled water. A raw (stock) solution containing zinc wash water of 866 mg/L was used in the experiment. The working samples consist of 100%, 50% and 25% of solution prepared by dilution with distilled water to required levels. The samples were used freshly from the ice cold deep freezer by dilution from the stock as and when required. Before the experimentation, the pH of the solution was maintained to be 6.3. Other pH conditions i.e., 6.0, 7.0 and 8.0 were obtained by using 0.1NH_2SO_4 and 0.1N sodium hydroxide solution. All measurements were carried out at ambient temperature. The removals of zinc in wash water in terms of current density, pH, supporting electrolyte variation were determined by electro coagulation. From the results percentage reduction of zinc was calculated. Spectral analysis of the samples was carried out using FT-IR, HPLC and SEM/EDAX.

The removal of zinc in wash water in terms of current density, initial concentration, Time was determined by electro coagulation for undergoing analytical processes the sample was given for analysis. HPLC, FTIR for understanding the extent of pollutant removal with reference to organic composition. The sharp and strong peak at 3442.8 and 3452 cm^{-1} was due to the O-H stretching vibration was observed from FTIR spectrum. In HPLC analysis, the efficiency for the electrochemical treatment is found as high as 99.9% shows the complete removal of zinc ions (zinc chloride). XPS spectrum indicates the presence of different phases. SEM image shows an aggregated formation of oxide and fine coagulant particles. EDAX spectrum shoes the presence of other elements in the sludge.

The experiments were conducted on the electro chemical cell from CECRI, Karaikudi. Three input variables namely Initial Concentration, Current Density and time were selected to optimize the zinc removal from the wash water in the electro coagulation process, the levels of the process variables have been shown in the Table 1. Experimental data used in this study was recorded from a serious of electro coagulation experiments values and the predicted values of ANN with the error between the experimental and predicted values are showed in the Table 2.

Table 1 Levels of Control Parameters

Control Parameters	**Levels**		
	-1	**0**	**1**
Initial Concentration (Ic)	216.5	433	866
Current Density (Cd)	0.1	0.2	0.6
Time (t)	2	5	15

Table 2Values of ANN predictions and Experimental error %

S.No.	**Initial Concentration (Ic)**	**Current Density (Cd)**	**Time (t)**	**Percentage of Zinc Removal Rate (ZRR)**		
				ANN	**EXP**	**Error**
1	216.5	0.1	2	87.87	83.32	-5.46
2	216.5	0.1	5	79.71	83.32	4.33
3	216.5	0.1	15	52.50	66.65	21.23
4	216.5	0.2	2	62.10	74.96	17.16
5	216.5	0.2	5	53.93	49.98	-7.91
6	216.5	0.2	15	26.72	0	0
7	216.5	0.6	2	29.64	49.97	40.69
8	216.5	0.6	5	21.47	0	0
9	216.5	0.6	15	-5.74	0	0
10	433	0.1	2	108.66	90.9	-19.54
11	433	0.1	5	100.50	90.2	-11.42
12	433	0.1	15	73.29	81.8	10.41
13	433	0.2	2	85.72	100	14.28
14	433	0.2	5	77.56	80	3.06
15	433	0.2	15	50.34	60	16.09
16	433	0.6	2	64.59	88.87	27.32
17	433	0.6	5	56.43	54.54	-3.47
18	433	0.6	15	29.22	0	0
19	866	0.1	2	101.29	100	-1.29
20	866	0.1	5	93.13	91.67	-1.59
21	866	0.1	15	65.92	75	12.11
22	866	0.2	2	84.02	72.7	-15.56
23	866	0.2	5	75.85	72.7	-4.34
24	866	0.2	15	48.64	54.54	10.82
25	866	0.6	2	85.56	76.92	-11.23
26	866	0.6	5	77.40	76.92	-0.62
27	866	0.6	15	50.18	61.54	18.45

In this investigation, A Neural Network Toolbox of MATLAB@ (R2013a) mathematical software package that having different tool boxes for different engineering discipline which was employed to predict the percentage of zinc removal from the barrel plating wash water.

ANN design accuracy always depends upon the finding of the best network architecture, number of neurons, and number of hidden layer, transfer functions, proper activation functions and training algorithm determine the adequate neural network topology of the problem and the basic principle of the ANN design as shown in figure 4.

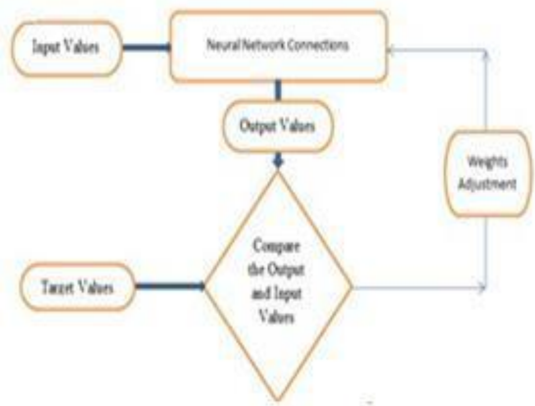

Figure 4 Basic Principle of ANN

Totally 27 experimental sets were used to develop the ANN model network which was trained automatically with the MATLAB software function "Train" with the "weights" and "biases" initialized in random values and randomly divided into three subsets: training (60% samples), validation (20% samples) and testing (20% samples) and the data were loaded into the workspace at random for each subset. Many network architectures were attempted. Based on the below equation (1), all the data (Xi) are converted into normalized value (Xnormal) and the normalized values are bounded in between Zero to one.

$$X_{normal} = (X_i - X_{min}) / (X_{max} - X_{min}) \quad ----- \quad (1)$$

Where X_{min} and X_{max} are represented as minimum and maximum actual experimental data.

The batch experiments data was separated into input matrix [p] and output matrix [z]. Based on the Mean Squared Error (MSE) and Correlation Coefficient (R^2) values of training and prediction set, the model and its variables were concluded and the performance of the ANN model is found with the minimum value of MSE and maximum value of R^2. Finally, the whole data sets were putted into the network and analyzed the regression between the network output and corresponding targets (response).

For Validation of model, the following statistical indices such as MSE and correlation coefficient (R^2) which are employed (N.Sathisha et al [13]) & (D.Sarala Thambavani [12]) respectively to evaluate the integrity of the fit of experimental data and the prediction accuracy of the model,

$$MSE = \frac{1}{Q}\sum_{k=1}^{Q} e(k)^2 = \frac{1}{Q}\sum_{k=1}^{Q}[t(k) - y(k)]^2 \quad ------ (2)$$

$$R^2 = \frac{\sum_{p=1}^{N}(t_p - t_{mean})^2 - \sum_{p=1}^{N}(t_p - o_p)^2}{\sum_{p=1}^{N}(t_p - o_p)^2} \quad ------ (3)$$

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Here e (k) the error between the target and ANN output, t(k) target output, y (k) ANN output value and Q is the number of total data. Where R^2 is the correlation coefficient, N is the number of the patterns, p is the index number for pattern, t_p is the target value for the p^{th} pattern, t_{mean} is the mean target value and O_p is the output of the pth pattern which is produced by the ANN model.

In the field of optimization, GA was used to: optimize the functions, process of images, solve trade man problem, identification systems and control and so on. GA is an evolutionary algorithm which applies the idea of survival of the fittest amongst an interbreeding population to create a robust search strategy and to find the best approximations. In general, the genetic algorithms are an excellent option for the global robust search of an optimal value from non-linear and high dimensionality functions. The optimization seeks to maximize or minimize the value of a function in a given search space. GA mainly works on three types of operators: reproduction, crossover and mutation. The data processed by GA are represented by an array of strings (chromosomes) with finite length. Where each bit is called allele or gene. The process conditions are encoded as genes by binary encoding to apply GA in optimization of processing parameters. Genes are represented with finite length of binary codes: 0 and 1. Chromosomes are the string of defining genes consists of 3 genes corresponding to 3 searching parameters(Ic, Cd and t). To evaluate each individual or chromosome, the encoded processing conditions are decoded from the chromosomes and a set of genes is combined together to form chromosomes, used to perform the basic mechanisms in GA, such as crossover and mutation. The crossover is the main operator, which generates new strings, eventually with better fitness values? Crossover operator aims to interchange the information and genes between chromosomes. Crossover is applied with probability which is normally set equal to 0.8.

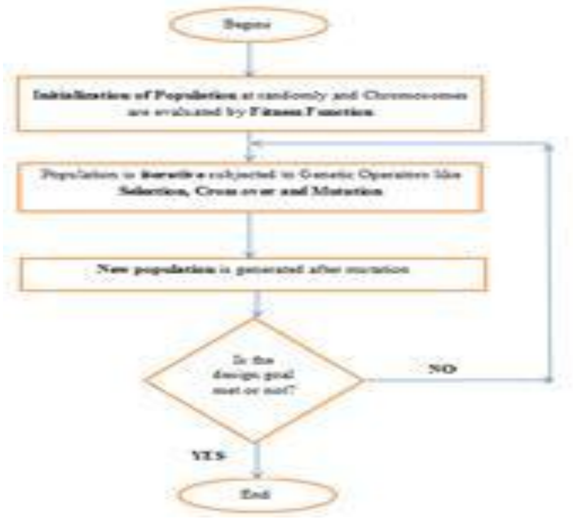

Figure 5 GA Flow Chart for Optimizing

After crossover, mutation is performed to ensure some randomness in the new chromosomes. Mutation operator aims to achieve some stochastic variability of GA in order to get a quicker convergence. Usually set to be small, normally 0.01. A value of the fitness function is attached to each individual, in order to evaluate its quality. A collection of strings is called population and the population at a certain point is referred as generation. The generation of the initial population of strings is done in a random way. Reproduction process goal is to allow the genetic information, store in the good fitness artificial strings and survive the next generation. Selection mechanism is applied to copy the survival members from the current generation to the next one. Selection is usually implemented through the so called process roulette wheel.

A program used to optimize the electro coagulation process parameter was created by implementing the flowchart given in figure 5. GA have three components namely initialization, reproduction and selection. Objective function is used for optimization as follows:

Maximize;

Function [z] = removal(x)

$$z = -(+ 91.88447 + 0.19598 * x_1 - 392.06786 * x_2 - 2.72117 * x_3 + 0.13089 * x_1 * x_2 - 1.74051E^{-004} * x_1^2 + 353.22222 * x_2^2);$$

end with the upper and lower levels given by;

$$216.5 \leq \text{initial concentration (mg/L)} \leq 866$$
$$0.1 \leq \text{current density (A/dm2)} \leq 0.6$$
$$2 \leq \text{time (minutes)} \leq 15$$

In optimization tool genetic algorithm solver is selected. Objective function obtained from the design of expert software use as a fitness function of GA tool. GA begins with a set of solutions represented by chromosomes, called population. Solutions from one population are taken and used to form a new population. This is motivated by the possibility that the new population will be better than the old one. Further solutions are selected according to their fitness to form new solutions (offspring's). The above process is repeated until some condition is satisfied. Trial and error method for the selection of initial population size found the best result when the size of 10 was chosen. The initial population size considered while running the GA is 10 and a test of 13 runs with 51 iterations each has been conducted and the outputs are listed in table 3 for maximize the zinc removal rate.

Table 3 Outputs from the Genetic Algorithm

S.No.	Concentration (Ic)	Current Density (Cd)	Time (t)	Zinc Removal Rate (ZRR)
1	668.03	0.19	2.03	94.66
2	355.69	0.14	3.27	90.34
3	534.52	0.15	2.13	100.36
4	410.52	0.1	5.49	97.78
5	504.64	0.11	2.43	109.47
6	667.41	0.15	5.25	92.67
7	428.91	0.1	3.42	104.33
8	217.5	0.1	2	88.01
9	698.54	0.1	7.96	95.65
10	544.02	0.23	6.72	73.97
11	645.72	0.6	3.51	78.94
12	810.03	0.6	2.29	85.73
13	480.27	0.2	2.45	87.45

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III. RESULTS AND DISCUSSIONS

Development of ANN model for Zinc removal

Predicted values of ANN and the experimental values for the zinc removal rate for the 27 input - output experimental training set are found by using C program as shown in the table 3.

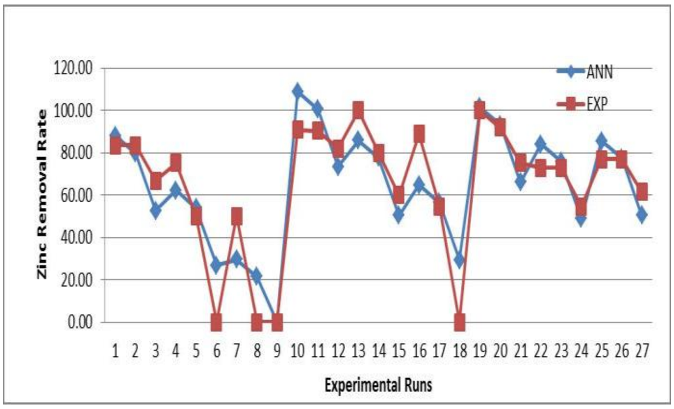

Figure 6 Comparison of ANN prediction with the experimental values of ZRR

The error can be calculated using equation below. The value of average error was obtained 10.31%. It is concluded that the ANN model can give adequate prediction of the zinc removal rate. Experimental zinc removal rate were compared with zinc removal rate predicted values of the ANN model is shown in the figure 6. It is noted that the figure indicates the prediction of zinc removal rate from the ANN model closely agree with that of experimental values. ANN model was developed automatically by the feed forward back propagation algorithm with Levenberg-Marquardt training with the mean squared error (MSE) and R^2 are used as performance functions. The network model having three layers such as input layer with three neuron (initial concentration, current density and time), output layer with one neuron (zinc removal rate) and one hidden layer with suitable number of neurons, for each neuron were analyzed.

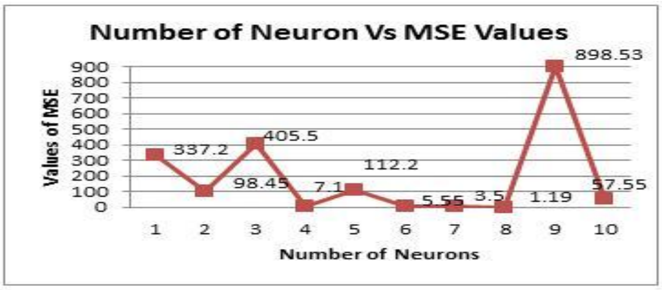

Figure 7 Number of Neuron

Figure7 shows the number of neurons in the one hidden layer. It was selected as 8 neurons because of its minimum error values (%) for training sets.

In this study, the optimal topology of the developed three-layered feed forward back propagation ANN architecture and the output of MATLAB software ANN model were obtained as shown in figures 8 & 9.

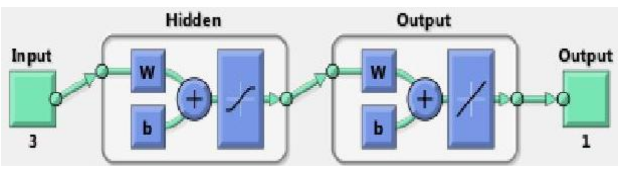

Figure 8 Topology

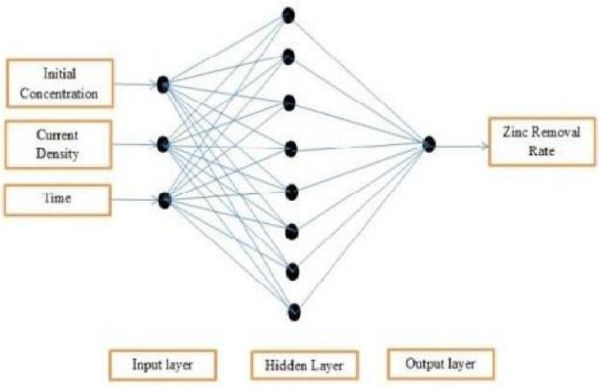

Figure 9 Structure

Performance of the network is determined with MSE which should be minimized and R^2 which should be maximized. Set the MSE (training epochs are continued until the MSE fell below this value) minimum value as 0.0001 and R^2 maximum value as 1, with the selective activation hyperbolic tangent function of hidden and output neurons.

The training, validation and testing mean squared error were illustrated in figures 10 & 11. When an epoch is equal to 8, the training was stopped. The performance of the regression analysis of proposed network response between ANN predicted output and the experimental targets with the graphical output of the network outputs plotted versus the targets as open circles were illustrated in the above Figures. From the figure 10, because of the non-linear dependence of the data, linear regression shows a good agreement between ANN outputs (predicted data) and the corresponding targets (experimental data).

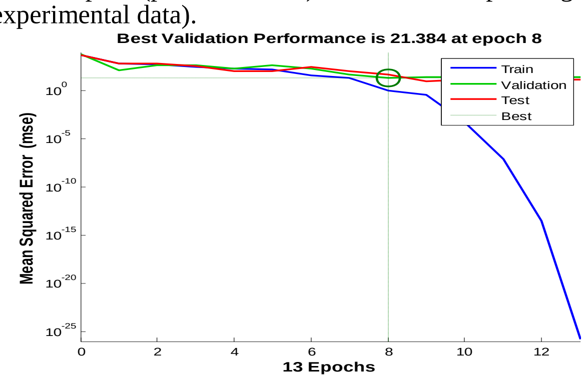

Figure 10ANN output

The prediction model was shown that the ANN model was able to predict the zinc removal rate from the waste water. These results are quite reasonable and accepted. The best linear fit was indicated by a solid red line and R^2 is almost 0.9909. Based on the value, the prediction accuracy of the ANN model is good. The mean squared error (MSE), correlation coefficient (R^2) and other statistical measures for training, validation, testing and all data sets are summarized in Table 4.

Table 4Performance of ANN model for Training, Testing, Validation and all data

Best Archi tecture	Correlation Coefficient (R2)				Mean Squared Error (MSE)			
	Train ing	Test ing	Valid ation	All	Train ing	Testi ng	Valid ation	All
3/8/1	0.999	0.98	0.979	0.99	1.19	21.38	48.72	23.8

The training errors, validation errors, and test errors for percentage removal plots of individual activities are shown in the Figure 11. The final mean square error, the test set error and the validations set error were very small. The best validation performance occurred by many iteration and the results observed were reasonable. It was observed that the predicted (ANNs) and experimental values were fairly following the similar trend for the removal of zinc.

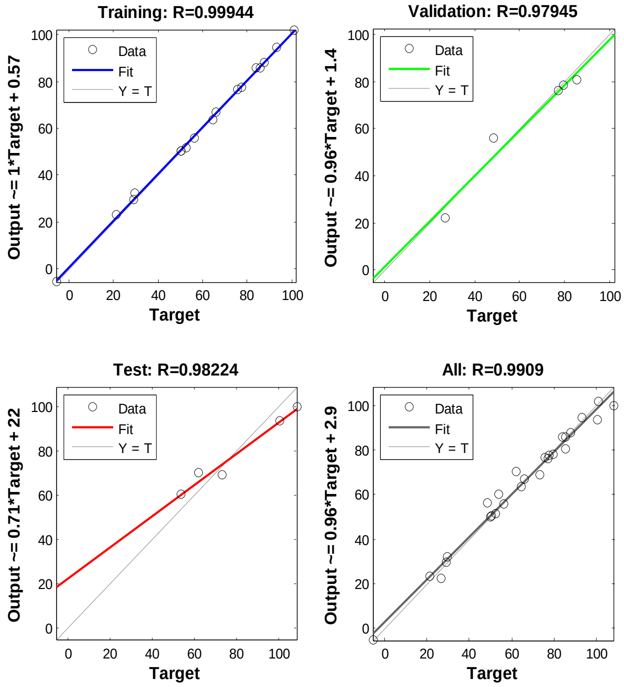

Figure 11 ANN error output

Optimization using Genetic Algorithm:

GA was used to optimize the control variables for zinc removal from the industrial wash water by using electro coagulation process. The table 5 shows the conditions and parameter constraints set for the optimization of zinc removal from the wash water using GA environment.

Table 5: GA Conditions and Constraints

Details of GA Solver	Values
Population	10
Population Type	Double vector
Scaling Function	Rank
Selection Function	Stochastic Uniform
Elite Count	2
Cross over fraction	0.8
Mutation Function	Adaptive Feasible
Crossover Function	Intermediate
Crossover Ratio	0.9
Direction	Forward
Level of Display	Iterative

From the figure 12, the best values of Best and Mean are detailed as 0.8745 and 0.8745 respectively.

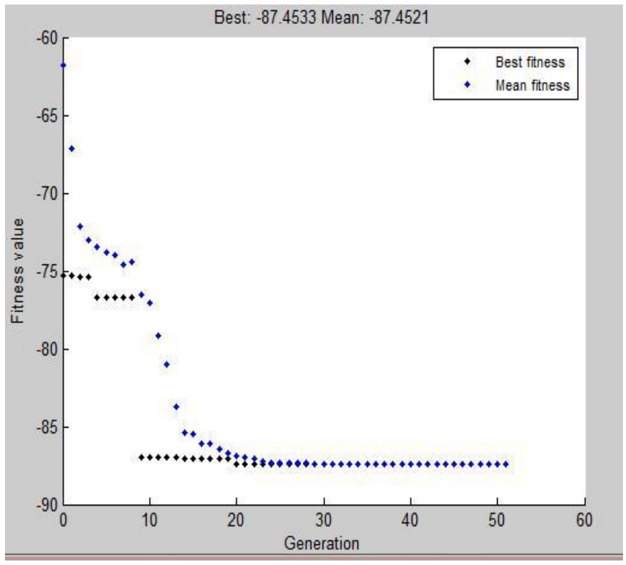

Figure 12GA graph

From the table 6, the optimum values of variables obtained for the removal of zinc are Initial Concentration Ic = 217.5 mg/L, Current Density Cd = 0.1A/dm² and TimeT = 2 mins and the maximum zinc removal percentage is 88.01%.

Table 6 Optimum Results from the Genetic Algorithm

Initial Concentration (Ic)	Current density (Cd)	Time (t)	Zinc Removal Rate (ZRR)
217.5	0.1	2	88.01

The conformation experiment for checking the accuracy of given quadratic model in the equation was conducted. The result details at optimum level as shown in Table 7.

% Error = (Residual / Actual Value) x 100% --- (4)

Table 7 Details of Conformation Test

Initial Concentration (Ic)	Current Density (Cd)	Time (t)	Value of Zinc Removal Rate (ZRR)		Error (%)
			Predicted	Experimental	
217.5	0.1	2	88.01	87.87	0.15

From the table 7, the percentage of error between the predicted and experimental values is 0.15% and the percentage of error is below 5% so the model is acceptable.

IV. Conclusions

The present research has discovered the application of ANN in modeling and GA in optimization of zinc removal from the wash water by using electro coagulation process. The optimal neuron number for the LMA was determined to be 8 hidden neurons with MSE of 1.18 and the proposed ANN model showed a precise and an effective prediction of the experimental data with a satisfactory correlation coefficient of 0.9909. ANN model based on back propagation algorithm proved to be very efficient in predicting the desired output (Zinc Removal Rate). The GA was focused on solving the optimization problem, where the removal rate of zinc was maximized. To improve the results, a special attention was given to the choice of the appropriate control variables of GA

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for the approached electro coagulation process. It was found that simulation produces the optimum zinc removal rate is about 88.01%. The optimum conditions for the three variables were obtained and compared with the real laboratory experiments, ascertaining a good agreement between them. Errors under minimum proved that the GA optimization is an efficient method.

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