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Predictive modeling of deposition rate in electro-deposition of copper–tin using regression and artificial neural network

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ABSTRACT

The aim of this paper is to develop a model using artificial neural network for the electro-deposition of copper–tin alloy (bronze) based on the experimentally obtained data. Copper–tin alloy was electrodeposited from a cyanide bath. The coating composition was determined using X-ray fluorescence spectroscopy. The deposition rate was calculated from the mass, composition and area of the deposit and its approximate density. The results were used to create a model for the plating characteristics using ANN. The ANN model was compared with the regression model for analysis.
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1. Introduction

The quest for newer materials is mounting with the industrial revolution and automation. Newer materials with unique properties which were thought impossible in yester years are emerging and produced in a mass level with ease and precision using simulation and modeling. With the help of a few experimental data on a lab scale, it can pin point the operating conditions to obtain coatings with the desired properties on a production level. Evidently this can be used to bring about property improvements in a variety of existing commercial products.

Copper–tin alloy, commercially known as tin–bronze, are stronger and more ductile and are usable at higher temperatures than the leaded alloys. Their high wear resistance and low friction coefficient against steel is useful in bearings, gears and piston rings. Mostly, they are produced as electrodeposits on steel substrates. To improve the lubrication properties further, attempts are being made to incorporate graphite or Polytetrafluoroethylene (PTFE) during electro-deposition of copper–tin alloy. Prior to the production of the improved lubricant coating, it becomes mandatory to fix the operating conditions to obtain the bronze alloy with the required composition, thickness and properties.

The effects of electroplating variables like current density, stirring rate and bath temperature on the properties of electrodeposits were discussed by Vernon et al. [1]. Coatings of copper–tin over mild steel substrate were carried out and the electroplating parameters were studied [2]. A regression model and artificial neural networks model have been developed by Ramanathan, for the prediction of hardness [3] and volume percent of diamond [4] in Ni–diamond composite coatings. The ANN prediction is reported to be closer to experimental values than that of regression model. Modeling with ANN was used for quantitative analysis of overlapped linear scan voltammetric and differential pulse polarographic peaks of adenine and cytosine that occur in the region of hydrogen evolution [5]. The average absolute error reported was a maximum of 5.9%. ANN was deployed as an effective theoretical tool to understand the charge–discharge characteristics of lithium–ion cells [6]. The correlation coefficient for the prediction of cycle life characteristics of Li–ion cell with CoO anode was reported as 0.98. Predictive modeling of copper in electro-deposition of bronze was carried out using regression analysis (SPSS 15 software) and ANN [7]. Cathode efficiency and deposition rate prediction in electro-deposition of bronze and prediction of tin content in the deposits were carried out using regression and ANN [8,9]. Numerous reports are available on the development of mathematical models relating process variables and bead geometry for the selection and control of the procedural variables [10–12] and prediction of tool life [13–18]. Models based on neural networks in predicting accurately both surface roughness and tool flank wear in finish dry hard

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turning have been developed [19] and the neural network models have been compared with the mathematical regression models.

It is reported that the neural network models have better prediction capabilities than regression model.

This paper deals with the development of a mathematical model for the prediction of deposition rate (DR) of copper–tin alloy coatings using MINITAB 15 software package. The results with particular reference to the electroplating parameters are also predicted using artificial neural networks and compared.

2. Experimental setup used for the present study

The electroplating setup used for the experiment, as shown in the Fig. 1, consists of a dual anode assembly viz., copper and tin, and a single mild steel cathode. Additionally the setup has a magnetic stirrer, current generators (rectifiers), and speed and temperature control devices. The distance between anode and cathode was 7 cm. The coating area was 6.25 cm^2. Bronze alloy of the required composition was produced from a cyanide electrolyte containing, CuCN-30 g l^{-1}; NaCN-45 g l^{-1}; Na_2SnO_3-42 g l^{-1}; NaOH-10 g l^{-1}; pH-12.5. The plating parameters were optimized by varying the current density from 1 to 5 A dm^{-2} (i), stirring rate (n) from 50 to 300 rpm and bath temperature (t) from 40 to 60 °C. The percentage of copper and tin content in the coatings was determined using X-ray fluorescence spectroscopy. The alloy deposition rate in μm h^{-1} was calculated using the formula given in

$$DR = \frac{\text{mass of the alloy deposit per hour}}{\rho_{\text{alloy}} \times \text{area}} \tag{1}$$

where, ρ_{alloy} is the alloy density which is given by,

$$\rho_{\text{alloy}} = \rho_{\text{Cu}} \times \text{mass percentage of copper} + \rho_{\text{Sn}} \times \text{mass percentage of tin}. \tag{2}$$

The observed values of i, n, t and DR are given in Table 1.

3. Development of regression model

The purpose of developing the regression model relating the deposition rate and electroplating parameters is to facilitate a functional relationship between deposition rate and the independent variables (current density, stirring rate and bath temperature). The response function representing the deposition rate of the coatings can be expressed as DR = $f(i, n, t)$ and the relationship selected was second-degree response. Out of 25 sets of experimental data available in Table 1, which contain different values of process variables and the corresponding experimental outputs, 20 have been taken for training and the remaining five sets of data were taken for validation. To establish the prediction model, regression analysis was carried out with MINITAB 15 software package, based on the method of least square for 95% confidence interval.

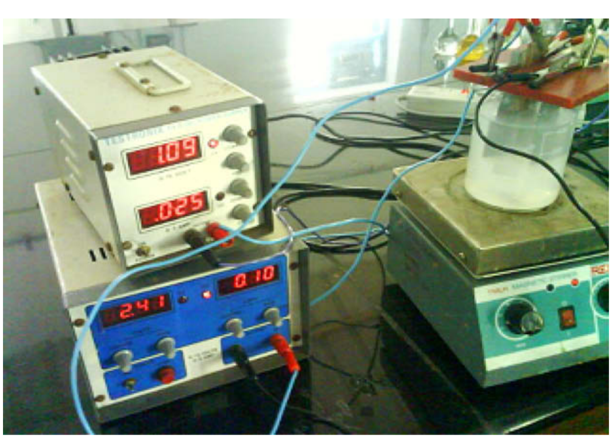

Fig. 1. Electroplating setup.

Table 1
Process variables and actual values of deposition rate.

Sl. No.	i/A dm^{-2}	n/rpm	t/°C	DR/μm h^{-1}
	Process variables			Actual values
1	1	50	60	17.00
2	2	50	60	29.53
3	3	50	60	39.48
4	4	50	60	47.32
5	5	50	60	47.76
6	1	80	40	18.32
7	2	80	40	32.98
8	3	80	40	45.24
9	4	80	40	54.20
10	5	80	40	51.61
11	1	300	40	19.60
12	2	300	40	35.87
13	3	300	40	50.45
14	4	300	40	58.03
15	5	300	40	59.71
16	1	300	50	18.90
17	2	300	50	35.05
18	3	300	50	49.23
19	4	300	50	55.83
20	5	300	50	52.46
21	1	300	60	17.83
22	2	300	60	30.30
23	3	300	60	40.62
24	4	300	60	51.72
25	5	300	60	49.40

The criterion to judge the efficiency and the ability of the model to predict the (DR) was taken as percentage error which is defined in

$$\%\text{Error} = \frac{(\text{Actual value} - \text{Predicted value})}{\text{Predicted value}} \times 100. \tag{3}$$

With this criterion it would be easier to understand how the regression model fits and to what extent the predicted values are close to the experimental values. The values of the regression coefficients give an idea as to what extent the control variables affect the responses quantitatively. The results of regression analysis with all the variables and most of the interaction terms in second-degree response and the corresponding coefficients and P-values are shown in Table 2.

The insignificant model term nt was automatically eliminated by the software since it is highly correlated with other variables. The most insignificant terms like temperature (t) and second order effect of temperature (t^2) were eliminated from the model, since their respective P-values in Table 2 are higher. The other less significant coefficients were dropped in the same manner, along with the responses with which they are associated, without sacrificing accuracy. The significant coefficients thus selected were recalculated and shown in Table 3. Comparing Table 2 and Table 3, it is evident that the removal of less significant terms have improved the R-Sq(adj) value from 0.982 to 0.984. The P-values in Table 3 for the independent variable, square term, and other interaction terms are below 0.05. Hence the main effect of current density (i), the second order effect of current density (i^2), the two level interactions of current density and temperature (it) and current density and stirring rate (in) are significant model terms. However, the main effect of current density (i) and the second order effect of current density (i^2) are the most important factors influencing DR.

The F-ratio from statistical table is 3.06 for a level of confidence of 95%. Referring to F-ratio of 292.47 in Table 3, which is greater

Table 2
Regression analysis of deposition rate for all terms.

Predictor	Coefficient	SE coefficient	T	P
Constant	0.1800000	30.47000000	0.01	0.995
i	27.0720000	2.39600000	11.30	0.000
n	−0.1173000	0.10310000	−1.14	0.279
t	0.1620000	1.21300000	0.13	0.896
*i***2	−2.6436000	0.27310000	−9.68	0.000
*n***2	0.0003330	0.00028250	1.18	0.263
*t***2	−0.0027300	0.01211000	−0.23	0.826
*i***n*	0.0028180	0.00246300	1.14	0.277
*i***t*	−0.0604800	0.03175000	−1.90	0.083
S = 1.97386	R-Sq = 98.9%		R-Sq(adj) = 98.2%	

Analysis of variance (ANOVA)

Source	DF	SS	MS	F	P
Regression	8	3970.37	496.30	127.38	0.000
Residual Error	11	42.86	3.90		
Total	19	4013.22			

SE: standard error; *T* (value of student's *t*-distribution): estimated coefficient/SE Coefficient; *P*: probability density; *S*: $\sqrt{MS\ \text{error}}$; R-Sq = 1-(SS Error/SS Total); R-Sq(adj):1-(MS Error/(SS Total/DF Total)); DF: degree of freedom; SS: sum of squares; MS: mean square; *F*(value of *F*-distribution): MS Regression/MS Error.

Table 3
Regression analysis of deposition rate for significant terms.

Predictor	Coefficient	SE Coefficient	T	P
Constant	−4.482000000	1.927000000	−2.33	0.034
i	27.693000000	1.722000000	16.09	0.000
*i***2	−2.670700000	0.253700000	−10.52	0.000
*i**n	0.003863000	0.001076000	3.59	0.003
*i***t*	−0.074180000	0.013860000	−5.35	0.000
S = 1.84040	R-Sq = 98.7%		R-Sq(adj) = 98.4%	

Analysis of variance

Source	DF	SS	MS	F	P
Regression	4	3962.42	990.60	292.47	0.000
Residual Error	15	50.81	3.39		
Total	19	4013.22			

than that of statistical table value, yields a statistically significant regression model. The final model was developed using only these significant coefficients and is given in

$$DR = -4.482 + 27.693i - 2.6707i^2 + 0.003863in - 0.07418it \quad (4)$$

R^2 and R^2(adj) values for the prediction of DR are 0.987 and 0.984, respectively. The validity of the equation developed is evident from their extremely high coefficients of correlation.

3.1. Checking the adequacy of regression model

A plot of the residuals vs. predicted (fitted) values for DR is shown in Fig. 2. The appearance of the horizontal band indicates that there is no violation of the model. Partial Least Squares (PLS) residual normal plot for 95% confidence level, for the predicted DR using four components in the response equation is shown in Fig. 3. The residuals for the plot of DR appear to follow a straight line and there is no evidence of non-normality, skewness and outlier [20]. All the points are inside the 95% confidence level which indicates that there is no problem with normality. Hence the developed regression model is adequate.

3.2. Validation of regression model

To test the accuracy of the model in actual applications, conformity test was conducted for the remaining five sets of data within

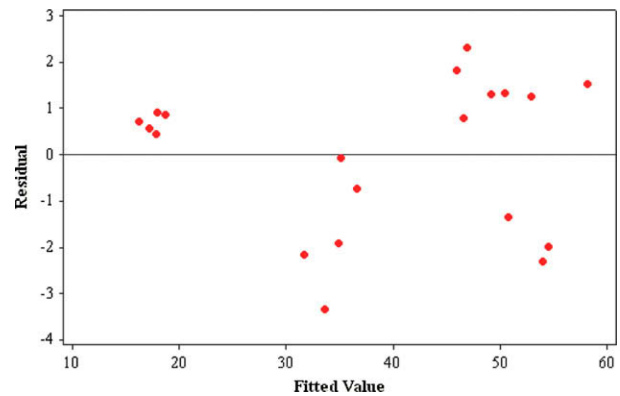

Fig. 2. Residual vs. fitted values for deposition rate.

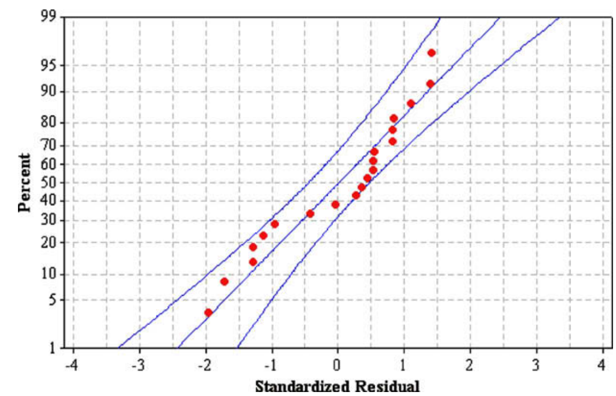

Fig. 3. PLS Residual normal plot for deposition rate.

the working limits for which different values of process variables and the corresponding experimental outputs are available. The percentage of errors, which give the deviation of predicted results of responses from the actual measured values, were also calculated and presented in Table 4. It is found from the table that the Mean Absolute Percentage Error (MAPE) for the regression model is 5.03%.

4. Artificial Neural Networks (ANN)

Matlab 7.0 is a software package used for high performance numerical computations and visualization. It provides an interactive environment with hundreds of built in functions for technical computations, graphics and animations. Built in functions provide excellent tools for linear algebra computation data analysis, signal processing, optimization and other scientific computations. In this work ANN module is utilized for predicting plating parameters for DR.

Neural networks are non-linear mapping systems that consist of simple processors, which are called neurons, linked by weighted connections. Each neuron has inputs and generates an output that can be seen as the reflection of local information that is stored in connections. The output signal of a neuron is fed to other neurons as input signals via interconnections. Since the capability of a single neuron is limited, complex functions can be realized by connecting many neurons. It is widely reported that structure of neural network, representation of data, normalization of inputs–outputs and appropriate selection of activation functions have strong influence on the effectiveness and performance of the trained neural network [21]. Methods such as Bayesian regularization and early stopping are commonly used to improve the

Table 4
Comparison of actual and predicted values of DR for validation data.

Sl. No.	i/A dm^{-2}	n/rpm	t/°C	DR/μm h^{-1}				
	Process variables			Predicted values from Regression	Predicted values from ANN	Actual values	Regression %Error	ANN %Error
1	3	50	60	41.79	40.53	39.48	−5.52	−2.60
2	3	80	40	46.59	45.12	45.24	−2.89	0.27
3	4	300	40	56.33	56.88	58.03	3.03	2.02
4	4	300	50	53.36	55.79	55.83	4.63	0.07
5	3	300	60	44.69	44.52	40.62	−9.10	−8.76
Mean Absolute Percentage Error (MAPE)							**5.03**	**2.74**

generalization in neural networks [22]. It is advantageous to use Bayesian regularization when there is limited amount of data [23]. Number of neurons to be used in the hidden layer of a neural network is critical in order to avoid over fitting problems, which hinders the generalization capability of the neural network. Number of hidden layer neurons is usually found with trial and error approach.

A neural network usually consists of three layers i.e., input layer, hidden layer and output layer, where inputs are applied at the input layer and outputs are obtained at the output layer and learning is achieved when the associations between a specified set of input–output pairs are established.

4.1. ANN parameters used for the study

There are different types of architecture for ANN mode. For creating a deposition model neural network requires different experimental data with regard to different plating parameters and plating performance. The same twenty sets of experimental data considered for obtaining a regression model were taken for training the artificial neural network. The inputs and outputs are normalized by,

$$X_i = \frac{X_i}{X_{\max}} \tag{5}$$

where, X_i is the value of a feature and $X_{\max}$ is the maximum value of the feature.

Normalized input data were fed to the system which in turn gives DR as output. The ANN architecture model used for the prediction of DR has three inputs, two hidden layers and single output. In this study, DR is predicated with a feed-forward back propagation multi-layer neural network as shown in Fig. 4.

A network structure 3-4-2-1 was chosen for DR prediction. In this network TRAINLM (Levenberg–Marquardt algorithm)was selected as training function, LEARNGDM was chosen for the adaption learning function and the default values of learning rate 0.01and momentum constant 0.9 was chosen for training. TANSIG transfer function was chosen for all three layers in the network.

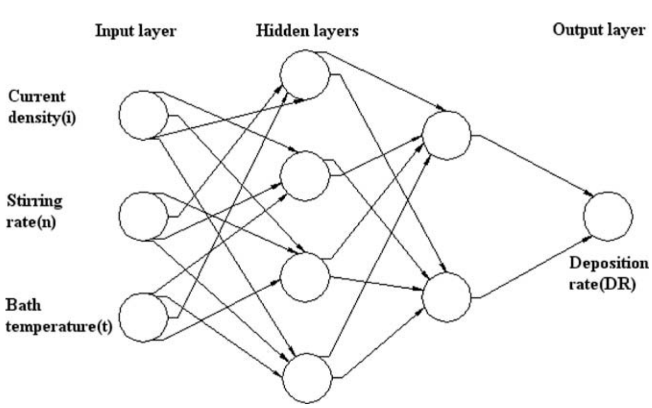

Fig. 4. Structure of a neural network.

The network's performance was measured according to the mean of squared errors (MSE). The training parameters like number of iterations, the acceptable mean-squared error (goal) and the adaptive value μ selected for the 3-4-2-1 network are shown in Fig. 5. The final weight and bias values to all the layers displayed by the network window which gives the functional relationship between input and output is given in Figs. 6 and 7, respectively. The performance curve for DR prediction is shown in Fig. 8. The performance of this network was compared with regression model.

4.2. Validation of neural network model

To test the accuracy of the 3-4-2-1 neural network model for the DR, validation test was conducted for the remaining same five sets of data within the working limits. The normalized outputs from neural network were converted to original form. The percentage of errors was calculated and presented in Table 4. It is found from the table that the MAPE for the ANN model is 2.74%.

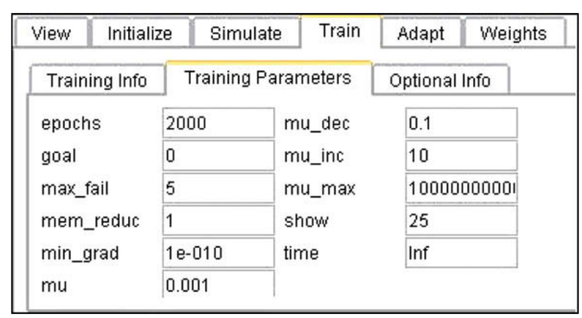

Fig. 5. Network training parameters.

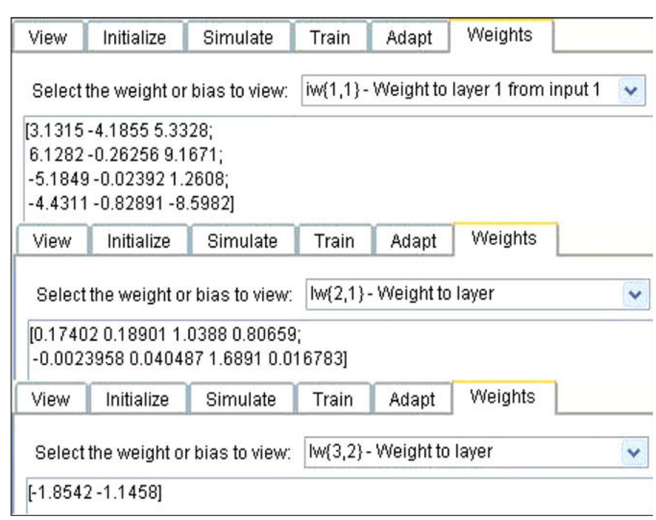

Fig. 6. Final weight to layers for 3421network.

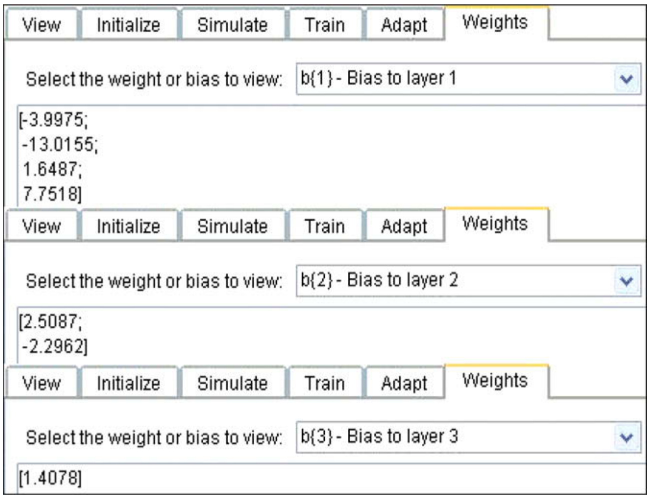

Fig. 7. Final bias to layers for 3421 network.

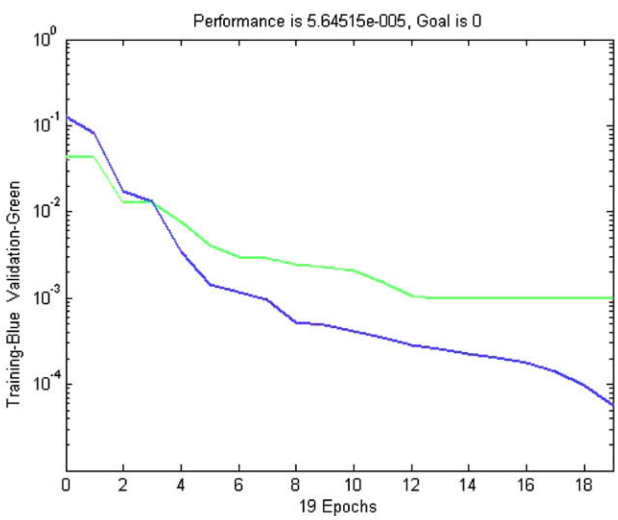

Fig. 8. Performance curve for deposition rate.

4.3. Comparison of prediction of deposition rate by regression and ANN

The deviation of predicted values from the actual values for the same twenty sets of training data for regression and ANN models were calculated and presented in Table 5. It is found from Table 5 that the MAPE for regression model is 3.80% and for ANN model is 0.92%. The DR predicted by both regression and ANN was compared with the experimental values for the same five sets of validation data and the results are presented in Table 4. While comparing the modeling accuracy for all the twenty five experimental data, it is found that the MAPE for regression model is 4.05% and that of ANN model is 1.28%. The comparison shows that ANN model is closer to experimental than that of the regression model.

5. Analysis of deposition rate using response surface methodology

Fig. 9 shows the surface plot of DR vs. stirring rate and temperature for a constant current density of 2 A dm^{-2}. DR increases with decrease in bath temperature and increases with stirring rate.

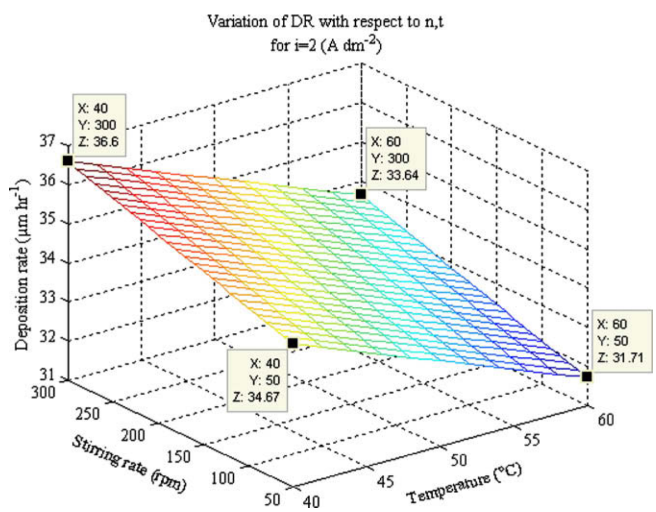

Fig. 9. Surface plot of deposition rate vs. stirring rate and temperature.

Table 5
Comparison of actual and predicted values of DR for training data.

Sl. No.	i/A dm^{-2}	n/rpm	t/°C	DR/μm h^{-1}				
	Process variables			Predicted values from Regression	Predicted values from ANN	Actual values	Regression %Error	ANN %Error
1	1	50	60	16.28	17.12	17.00	4.41	−0.71
2	2	50	60	31.71	29.20	29.53	−6.86	1.14
3	4	50	60	46.53	47.40	47.32	1.70	−0.17
4	5	50	60	45.93	48.37	47.76	3.99	−1.26
5	1	80	40	17.88	18.39	18.32	2.45	−0.38
6	2	80	40	34.90	33.02	32.98	−5.51	−0.11
7	4	80	40	52.93	54.08	54.20	2.41	0.22
8	5	80	40	53.92	51.50	51.61	−4.29	0.21
9	1	300	40	18.73	19.48	19.60	4.63	0.63
10	2	300	40	36.60	36.45	35.87	−2.01	−1.60
11	3	300	40	49.14	50.37	50.45	2.67	0.15
12	5	300	40	58.17	59.13	59.71	2.64	0.98
13	1	300	50	17.99	18.78	18.90	5.06	0.62
14	2	300	50	35.12	33.79	35.05	−0.20	3.74
15	3	300	50	46.91	49.48	49.23	4.94	−0.51
16	5	300	50	54.47	53.08	52.46	−3.68	−1.16
17	1	300	60	17.25	18.19	17.83	3.37	−1.96
18	2	300	60	33.64	31.07	30.30	−9.92	−2.49
19	4	300	60	50.39	51.61	51.72	2.64	0.21
20	5	300	60	50.76	49.32	49.40	−2.67	0.17
Mean Absolute Percentage Error (MAPE)							**3.80**	**0.92**

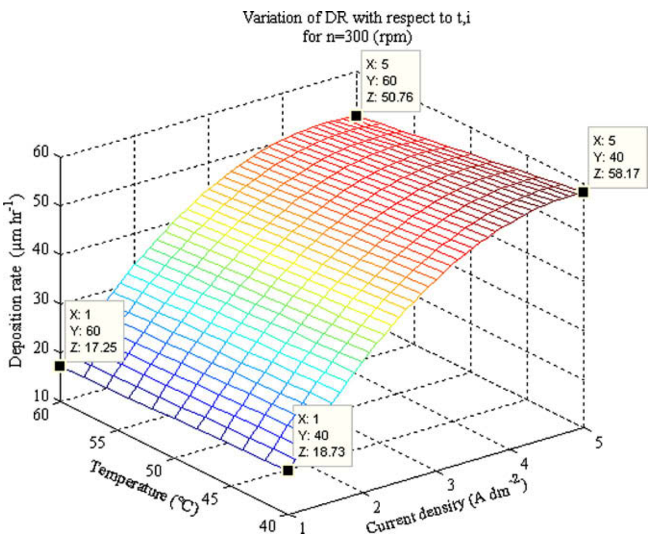

Fig. 10. Surface plot of deposition rate vs. temperature and current density.

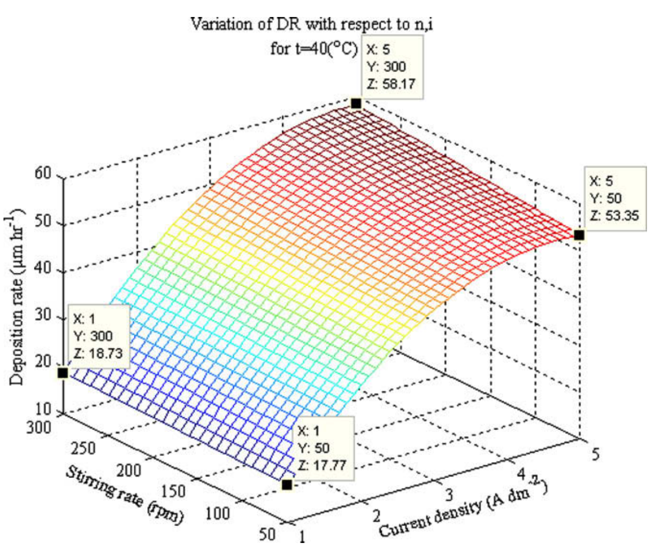

Fig. 11. Surface plot of deposition rate vs. stirring rate and current density.

Fig. 10 shows the variation of DR vs. temperature and current density for a constant stirring rate of 300 rpm. From the figure it is observed that DR increases with increase in current density and slightly decreases with bath temperature.

The surface plot of DR vs. stirring rate and current density for a constant bath temperature of 40 °C is shown in Fig. 11. It is evident from the Fig. 11 that DR increases with increase in current density and slightly increases with stirring rate.

6. Conclusion

The developments of model based on feed-forward back propagation network in predicting accurately, the DR of the electroplated specimens are carried out. The experimental data of measured DR is utilized to train the neural network model. Trained neural network model is used in predicting DR for various operating conditions. The developed system is found to be capable of accurate DR prediction for the range it has been trained. From the regression model it is interpreted that the main effects current density (i), the second order effect of current density (i^2) are the most significant model terms associated with DR prediction. The neural network model is also compared with the regression model. The neural network model provided better prediction capabilities because they generally offer the ability to model more complex non-linearties and interactions than linear and exponential regression model.

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