Application of feed forward neural network model to predict the limiting current of tin magneto electrodeposition

Sudibyo¹*, N Aziz², A Wijaya³ and R S Fathona³

¹Mineral Technology Research Unit, Indonesian Institute of Sciences, Lampung Indonesia
²School of chemical engineering, University of Science Malaysia, Penang, Malaysia
³Informatics Engineering, Sumatra Institute of Technology, Lampung, Indonesia

* E-mail: sudibyo@lipi.go.id

Abstract. Predicting the value of Tin Magneto electrodeposition (MED) is very important since the optimum mass transport occurred at the limiting current. The MED limiting current able to detect using electroanalytical chemistry, but this method is expensive; it needs some method, which able to predict the limiting current of tin MED. However, predicting the limiting current under magnetic field effect is more complicated due to the highly nonlinear characteristic and complicated of its multiple inputs single-output (MISO) system. The nonlinear model that able to predict the limiting current of tin MED is Artificial Neural Networks (ANNs). One of the ANNs which able to simulate the Multiple-Input-Single-Output (MISO) model is the Feed Forward Neural Network (FFNN). In this work, MISO FFNN will model a matrix data set with six variable inputs and one output. The data was obtained from the results of the experiments using electroanalytical chemistry. The output of this model is the limiting current of tin MED, meanwhile, the inputs are by the concentration of tin (Sn²⁺) in the electrolyte (C), viscosity(v), diffusion coefficien (D), area of the electrode (A), the number of electroactive species (n) and magnetic field strength (B). To get the best model, the performance of FFNN was tested with three variations of the algorithm (Levenberg-Marquardt, Bayesian Regularization, and Scaled Conjugate Gradient) and ten variations of the number of neurons (10, 15, 20, 25, 30, 35, 40, 45 and 50). The best model obtained for this MISO FFNN model is which uses the Levenberg-Marquardt algorithm and the highest number of neurons (50 neurons).

1. Introduction

Tin electrodeposition has been applied to microelectronics as an alternative for tin/lead finishes [1]. This process has a problem to obtain a uniform, dense, and compact deposition. Numerous studies have been addressed to reduce it. One of the methods available to solve is by adding a magnetic field in the electrodeposition process, this process known as magneto electrodeposition (MED) [2–4]. This MED technology has a great impact on the electrodeposition process to synthesize metal alloy, thin-film, multilayer, nanowires, multilayer nanowires, dot array, and nanocontacts which are the technology of the future to build the next generation of microelectronics devices [2].

The magnetic field effect caused the increase of mass transport on an electrochemical reaction in MED which indicated by the increase of limiting current. The optimum mass transport occurs at the limiting current, if the process conducted over the limiting current, this will cause the excess of hydrogen evolution reaction which will damage the surface quality of electroplating. Hence, knowing the limiting current (ib) is very important both in conventional electrodeposition processes or MED. Many
researchers have studied that the limiting current in MED is affected by many parameters such as the magnetic field strength (B), the number of electrons of the redox process (n), the electrode area (A), the diffusion coefficient of the electroactive species (D), the concentration of the electroactive species (C_{bulk}); and the kinematic viscosity of the electrolyte (v) [2].

The MED limiting current able to detect using electroanalytical chemistry. Since the electroanalytical chemistry is expensive and only in small scale (lab. Scale); it needs some method, which able to predict the limiting current of tin MED in the large-scale process. However, the limiting current under magnetic field (ib) is a very highly nonlinear process and complicated process which has multiple inputs with a single output. The nonlinear model that able to predict the limiting current of tin MED is Artificial Neural Networks (ANNs). One of the ANNs which able to simulate the Multiple-Input-Single-Output (MISO) problem is the Feed Forward Neural Network (FFNN). FFNN model also reported has been successfully simulated many nonlinear models such as temperature and pressure in the distillation and reactor process [5–7].

In this work, MISO FFNN will model a matrix data set with six variable inputs and one output. The data were obtained from the results of the experiments using linear sweep voltammetry (LSV) technique. LSV technique is one of the electroanalytical chemistry methods, which use potentiostat-galvanostat and three-electrode electrochemical reactor as shown in Figure 1. Platinum electrodes were used as a working electrode (WE) and counter electrode (CE), meanwhile, Ag/AgCl was used as a reference electrode (RE) in the three-electrode electrochemical reactor as shown in Figure 1.

The output of MISO - FFNN model is the limiting current of tin MED, meanwhile, the inputs are by the concentration of tin (Sn^{2+}) in the electrolyte (C), viscosity (v), diffusion coefficient (D), area of the electrode (A) and magnetic field strength (B). To get the best model, the performance of FFNN was tested with three variations of the algorithm (Levenberg-Marquardt, Bayesian Regularization, and Scaled Conjugate Gradient) and ten variations of the number of neurons. The FFNN model was generated using MATLAB software. The best model of MISO FFNN will be developed in the Simulink model which easy and simple to use.

![Image of an electrochemical cell](image-url)

Figure 1. Schematic illustration of an electrochemical cell.

2. Research methodology

2.1 Experimental work of Magneto electrodeposition (MED) Process in Tin

Tin (II) sulfate (SnSO_{4}) (≥99%, from Merck Co. Ltd.), Natrium sulfate (Na_{2}SO_{4}) (≥99% from Merck Co. Ltd), and Sodium Gluconate (≥99.8%, from R & M Co. Ltd.) are ingredients used in previous studies. All ingredients are prepared with the concentration determined using distilled water. The
magnet used in this research is a water-cooled superconductor electromagnet (Lake Shore EM 4, USA) with a 5 cm diameter pole and 5 cm distance. The intensity of the magnetic field is adjusted from 0 to 0.3 T. The analytical electrochemical techniques used are linear-sweep voltammetry (LSV) and Choromperometry (CA) using Potentiostat/ Galvanostat. In this work, LSV was used to measure the limiting current, meanwhile, CA was used to measure the diffusion coefficient of electroactive. The LSV results are then used as output in the FF NN modeling.

2.2 Development of the FFNN MISO Model
Feedforward neural networks (FFNN) develop from the approach of input and output data relationships from the existing experimental results. FFNN in this study will use 6 inputs, namely Tin concentration (M), electrode area (A), electroactive diffusion coefficient (D), kinematic electrolyte viscosity (v), magnetic field strength (B) and some electrons involved in redox process (n), from the 6 inputs will produce 1 output that is the limiting current value. To see the real results of each influence given this study uses 6 inputs on FFNN of Tin MED. Then, the entire input data is formed 6 x 1 matrix, while the output is in the form of limiting current (ib) in the form of a 1x1 matrix in Matlab. In this work, 120 data are divided into three parts, namely train (40%), validation (30%), and test (30%).

The structure of the neural network model consists of the input layer, hidden layer, and output layer network. The hidden layer functions, which consist of many neurons as a controller of complex estimations performed by non-linear transfer functions. The identification activity is ended until the best model is obtained from the simulation[5–7]. In this study, the identification command is typed in the m-file in Matlab. Simulations with MISO FFNN have varied the neurons from 10, 15, 20, 25, 30, 35, 40, 45, and 50 neurons and with 3 variations of the algorithm used namely, Levenberg-Marquardt, Bayesian Regularization, and Scaled Conjugate Gradient. The performance of the FFNN model was analyzed using mean square error (MSE). The best model is then imported into Simulink on Matlab.

3. Results and Discussion
The performance of MISO FFNN using various neurons and three variations of the algorithm used, namely, Levenberg-Marquardt, Bayesian Regularization, and Scaled Conjugate Gradient were listed in Table 1. Table 1 shows that the best performance of MISO FFNN using 50 of neurons and Levenberg-Marquardt of algorithm training with the smallest of MSE value. Figure 2 shows the validation performance of FFNN using the algorithm of Levenberg-Marquardt and 50 neurons. The figure shows that the model has MSE of 5.13 for training, 7.78 of MSE for validation, and 2.85 of MSE for the test of the neural network model. The suitable neuron number will give the best performance of the neural network. In this case, the highest number of neurons gives the best performance to simulate the limiting current of tin MED. The increasing of neurons numbers will increase the accuracy, but it will increase the time consuming for calculation. In this work, the accuracy is more important than the higher calculation time, hence this result is applicable for the limiting current prediction of tin MED.

![Figure 2. Validation performance of FFNN using the algorithm of Levenberg-Marquardt and 50 neurons.](image-url)
3.1 The Development of MISO FFNN in the Simulink Model.

The performance test of the model shows that MISO FFNN using the Levenberg-Marquardt algorithm with 50 neurons provides the best performance in modeling the Tin MED process. This model is written in the form of m-files which are then exported into Simulink in Matlab. The Simulink model was developed to simplify the use of MISO FFNN to predict the limiting current of tin MED as shown in Figure 3 [8]. This Simulink model has six inputs, namely Tin concentration (M), electrode area (A), the diffusion coefficient of electroactive (D), kinematic electrolyte viscosity (v), magnetic field strength (B) and the number of electrons involved in the redox process (B) n). The six inputs will be processed by FFNN and will produce an output in the form of a limiting current value (ib).

| Algorithm          | Neurons | TRAINING (MSE) (40% of Data) | VALIDATION (MSE) (30% of Data) | TESTING (MSE) (30% of Data) |
|--------------------|---------|------------------------------|-------------------------------|-----------------------------|
| Levenberg - Marquardt | 10      | 3.05                         | 5.84                          | 7.74                        |
|                    | 15      | 3.82                         | 5.95                          | 5.95                        |
|                    | 20      | 6.79                         | 3.40                          | 6.42                        |
|                    | 25      | 3.06                         | 8.98                          | 5.86                        |
|                    | 30      | 3.41                         | 7.60                          | 6.87                        |
|                    | 35      | 5.65                         | 4.10                          | 5.53                        |
|                    | 40      | 3.82                         | 11.50                         | 4.67                        |
|                    | 45      | 5.53                         | 4.10                          | 5.53                        |
|                    | 50      | 5.13                         | 7.78                          | 2.85                        |
| Bayesian Regularization | 10      | 5.15                         | 0.00                          | 5.94                        |
|                    | 15      | 4.40                         | 0.00                          | 7.55                        |
|                    | 20      | 6.28                         | 0.00                          | 2.95                        |
|                    | 25      | 5.55                         | 0.00                          | 4.64                        |
|                    | 30      | 5.60                         | 0.00                          | 4.44                        |
|                    | 35      | 4.86                         | 0.00                          | 7.41                        |
|                    | 40      | 5.58                         | 0.00                          | 4.60                        |
|                    | 45      | 6.20                         | 0.00                          | 3.44                        |
|                    | 50      | 4.86                         | 0.00                          | 6.16                        |
| Scaled Conjugate Gradient | 10      | 0.157                        | 13.90                         | 22.60                       |
|                    | 15      | 0.150                        | 16.80                         | 25.50                       |
|                    | 20      | 0.163                        | 16.80                         | 9.23                        |
|                    | 25      | 0.169                        | 18.00                         | 19.70                       |
|                    | 30      | 10.30                        | 17.60                         | 14.50                       |
|                    | 35      | 9.84                         | 10.10                         | 8.00                        |
|                    | 40      | 8.06                         | 12.20                         | 16.30                       |
|                    | 45      | 9.06                         | 12.20                         | 9.85                        |
|                    | 50      | 10.90                        | 11.80                         | 12.20                       |
3.2 The Effect of Tin Concentration towards the Limiting Current of Tin Magneto Electrodeposition

The second experiment was carried out by predicting the value of the limiting current (ib) by the various Tin concentration (M) using the FFNN model. The concentration of tin given was varied from 0.01 to 0.1 M and obtained the MSE of the limiting current value ratio (ib) data origin and value of the limiting current (ib) the data FFNN of 3.6302 x 10^{-6} as shown in Figure 4.

3.3 The Effect of Electrode Area towards the Limiting Current of Tin Magneto Electrodeposition

The third experiment was carried out by predicting the value of the limiting current (ib) by electrode area variation (A) using the FFNN model that had been obtained. Given the wide variation electrode is from 0.5 to 0.953 cm² and obtained the MSE of the limiting current value ratio (ib) data origin and value of the limiting current (ib) the data FFNN of 29.3542, shown in Figure 5. The error value for the small area of the electrode is big, hence this model is not good enough to simulate for a small area.
3.4 The Effect of Supporting Electrolytes Concentration towards the Limiting Current of Tin Magneto Electrodeposition

To find out the effect of supporting electrolytes on current limiting, the concentration of supporting electrolytes (Na$_2$SO$_4$) which varies from 0.075 to 1 M. The increase of supporting electrolyte concentration leads to the decreasing of diffusion coefficient. This phenomenon caused by the increase of electrolyte kinematic viscosity as the increment of supporting electrolyte concentration. Comparison between simulation results using the MISO FFNN model (identification using the Levenberg-Marquardt algorithm model with 50 of neurons) with original data (variation of diffusion coefficient as a function of increasing of supporting electrolyte concentration) gives the MSE value of 0.95113 as shown in Figure 6. The result shows that the error value is not suitable for the highest value of supporting electrolyte concentration.

3.5 The Effect of Additive Electrolytes Concentration towards the Limiting Current of Tin Magneto Electrodeposition

The effects of the current limiting electrolyte additive ($i_b$) for tin MED are studied by varying the gluconate shown in Figure 7. The increase of additive electrolytes will be caused by the decrease of limiting current ($i_b$). The decrease in the limiting current leads to increased friction as the viscosity electrolyte increases which reduces the magnetohydrodynamic (MHD) convection of the solution [2].
Although the presence of an additive will decrease the limiting current, the additive is useful for produce better electrodeposits such as to avoid a roughness on the metal surface, to reduce corrosion [9], to increase the purity of electrodeposits. Figure 7 also shows the MSE value of the experimental \( i_b \) towards the \( i_b \) value of FFNN gives very small of MSE value (0.00023015). This proves that this model has a good capability to predict \( i_b \) in different kinematic viscosity.

Figure 7. Comparison between simulation results using the MISO FFNN model (identification using the Levenberg-Marquardt algorithm model with 50 of neurons) with original data (variation of kinematic viscosity as a function of increasing of gluconate concentration).

3.6 The Effect of Magnetic Strenght (B) towards the Limiting Current of Tin Magneto Electrodeposition

Figure 8 shows the effect of a magnetic field variation (B) towards the value of the limiting current (\( i_b \)), in which the FFNN model able to predict the limiting current from the real experiment. The variation of the magnetic field (B) given is from 0.05 T - 0.3 T and the MSE value is obtained from the comparison of the limiting current value (\( i_b \)) of the experimental data with the FFNN result is 3.2877 \( \times 10^{-5} \). The result shows that the FFNN model is has a good performance to predict the \( i_b \) at the varying magnetic field.

Figure 8. The comparison of the limiting current model FFNN using (the model algorithm Levenberg - Marquardt and 50 of neurons) in predicting the limiting current toward the variations of magnetic fields.

3.7 The Effect of the number of electrons involved in the Redox Process (n) towards the Limiting Current of Tin Magneto Electrodeposition

The effect of the number of electrons involved in the redox (n) process on limiting the current of tin MED was studied using various supporting electrolytes which has different electron transfers such as Na\(_2\)SO\(_4\) (Na\(^{2+}\) has two-electron transfer), HCL (H\(^+\) has one electron transfer). The effect of supporting
electrolyte that has a single electron transfer was studied by a varied of HCl concentration from 1 – 3 M as shown in Figure 9. The figure shows the comparison of the limiting current model FFNN using (the model algorithm Levenberg - Marquardt and 50 of neurons) in predicting the limiting current toward the variations of HCl concentration gives MSE value of 0.00040895. This also proves that FFNN also suitable to simulate ib of tin MED using a variation of HCl.

4. Conclusion
The MISO FFNN model has successfully developed using Simulink on Matlab to predict the limiting current value of Tin MED and user friendly. The results show that the best performance of the FFNN model was achieved when the model generated using 50 neurons and the Levenberg-Marquardt algorithm. Several tests with a different value of input parameters (C, A, D, v, B, and n) shows that the FFNN model can predict the limiting current of tin MED.

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