Artificial Neural Network for Composite Hardness Modeling of Cu/Si Systems Fabricated Using Various Electrodeposition Parameters

I. Mladenović, J. Lamovec, V. Jović, M. Obradov, K. Radulović, D. Vasiljević Radović, and V. Radojević

Abstract –Copper coatings are produced on silicon wafer by electrodeposition (ED) for various cathode current densities. The resulting composite systems consist of 10 μm monolayered copper films electrodeposited from sulphate bath on Si wafers with sputtered layers of Cr/Au. Hardness measurements were performed to evaluate properties of the composites. The composite hardness (H_c) was characterized using Vickers microindentation test. Then, an artificial neural network (ANN) model was used to study the relationship between the parameters of metallic composite and their hardness. Two experimental values: applied load during indentation test and current density during the ED process were used as the inputs to the neural network. Finally, the results of the composite hardness (experimental and predicted) were used to estimate the film hardness (H_f) of copper for each variations of the current density. This article shows that ANN is an useful tool in modeling composite hardness change with variation of experimental parameters predicting hardness change of composite Si/Cu with average error of 6 %. Using created ANN model it is possible to predict microhardness of Cu film for current density or indentation load for which we do not have experimental data.

I. Introduction

Artificial neural network (ANN) is a numerical model designed to simulate information processing of a human brain. They are used in complex non-linear systems using the preexisting empirical data to learn about the system. As such ANNs are used for assessment, prediction, decision making and diagnostics [1,2]. The neural network consists of simple processors, called neurons. Each neuron has inputs and generates output signals that are sent to other neurons in the network as inputs via the interconnections. ANN approach is used in many fields of chemical and material engineering such as: prediction of yield strength, tensile strength and elongation of cast alloys [3], for estimation on of the deposition rate of copper-tin during electroplating, hardness predictions of nickle-CBN composites [2], evaluating the change of wood hardness during heat treatment [4], etc.

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Electrodeposited copper films are used in fabrication of micro-electro-mechanical (MEMS) devices for a wide range of applications [5]. Mechanical properties of electrodeposited copper films on silicon substrates in electronic devices heavily influence the lifetime of the devices [6]. As such it is especially important to analyze the hardness of the composite systems which depends on several factors, such as the microstructure and hardness of the film and of the substrate, thickness of the film etc. [7].

Microindentation is one of the best known methods for the evaluation of mechanical properties of films and coatings. In cases where the thickness of the film is small, the substrate hardness affects the hardness of the film, such a measured hardness is called composite hardness.

Based on experimental measurements, the database was created. Using that database we created neural network model that we used to predict composite hardness of our Cu/Si system. The measurements and predicted values of composite hardness were used to calculate the hardness of the copper film.

II. Artificial Neural Network (ANN)

In this study, a proposed ANN model was designed using the Matlab Neural Network Toolbox and using a multi-layer perception (MLP) model for prediction. The MLP architecture consists of an input layer, one or more hidden layers, and an output layer [4]. The input layer consists of two input nodes: applied load during indentation measurement and applied current density during ED process. The hidden layer utilizes three neurons, and the output layer consists of one output node: composite hardness of the Cu/Si systems.

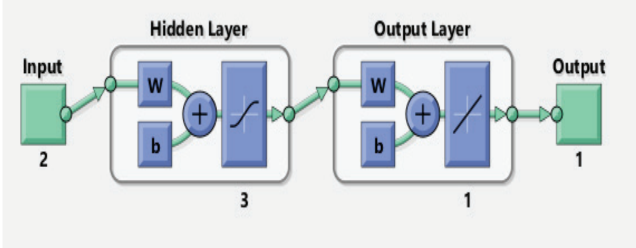

Fig. 1. Block diagram of ANN used in this study.

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ANN block diagram used in this study is given in Fig. 1. The hidden layer uses a hyperbolic tangent sigmoid transfer function and the training algorithm is the Levenberg-Marquardt backpropagation.

The database-containing 60 indentation hardness measurements of the fabricated Cu/Si composites was randomly divided into three groups: 42 data points (70% of the total data) used for the ANN training process, 9 data points (15 % of the total data) for validation group, and 9 data points (15 % of all data) for the testing process.

The network performance can be estimated through the error of deviation between actual and predicted values. The mean absolute percentage error (MAPE), the mean square error (MSE) and determination coefficient (R^2) were utilized to evaluate the performance of the ANN. The errors were calculated using the following formulas:

$$MAPE = \frac{1}{N}\left\{\sum_{i=1}^{N}\left[\left|\frac{Hc_i - Hcp_i}{Hc_i}\right|\right]\right\} \times 100 \quad (1)$$

$$MSE = \frac{1}{N}\sum_{i=1}^{N}(Hc_i - Hcp_i)^2 \quad (2)$$

$$R^2 = 1 - \frac{\sum_{i=1}^{N}(Hc_i - Hcp_i)^2}{\sum_{i=1}^{N}(Hc_i - Hc')^2} \quad (3)$$

where Hc_i represent the experimental output, Hcp_i represent the predict output, N represent the total number of samples and Hc' represents the mean of predicted outputs.

III. Hardness Determination of Copper Films Using the Work-of-Indentation Model

Mathematical models used to calculate the hardness of the thin film from the measured composite hardness depends on the type of composite systems. The composite hardness model of Korsunsky [8] was chosen and applied to experimental and prediction data in order to calculate the copper film hardness. According to this model correlation between composite hardness H_c, film hardness H_f, and substrate hardness H_s is given as:

$$Hc = Hs + \left[\frac{1}{1 + k' \cdot (d^2 / t)}\right] \cdot (Hf - Hs) \quad (4)$$

$$k' = \frac{k}{49 \cdot t} \quad (5)$$

where k is a dimensionless materials parameter related to the composite response mode to indentation, d is the indent diagonal and t is the thickness of the film.

Prior deposition process, the substrate hardness was first determined, experimentally. We used Proporcional Specimen Resistance (PSR) model [9]. The calculated hardness of the substrate was 14.65 GPa, measured for the first 5 load points.

IV. Experimental Procedure

For these experiments substrate of Si wafers (4 inch, (100) orientation) was chosen and prepared. The wafer was cut in parts about 1cm wide, standard cleaning and drying procedures. The plating base on the silicon wafers were sputtered layers of 10 nm Cr and 100 nm Au.

Copper films were electrodeposited from a 100 ml sulphate bath [6]. Electrochemical deposition was carried out using direct current galvanostatic mode with the 5 current density values (10, 33.33, 50, 66.67 and 100 mA/cm^2). Based on the platting surface, current density, and duration of deposition process, thickness of copper deposits was 10 μm.

The mechanical properties of the composite systems Si/Cr/Au/Cu were characterized using Vickers microhardness tester "Leitz, Kleinharteprufer DURIMET I" with loads ranging from 2.4515 N down to 0.04903 N. Indentation was done at room temperature. The dwell time was 25 s. The average values of impression diagonals d (in m), were calculated from several independent measurements on every specimen for different applied loads, P (in N). The composite hardness, Hc (in GPa), was calculated using the formula (6).

$$Hc = \frac{0.01854 \cdot P}{d^2} \quad (6)$$

Topographic examination was done by the metallographic microscope "Carl Zeiss Epival Interphako".

V. Result and Discussion

In this section, experimental results were compared to prediction values for composite hardness from the ANN model as shown in Fig. 2. Large disagreements between experimental and predicted values were noted for low and high loads. At low loads, the Vicker's diagonal size is small and difficult to read. These errors are known as indentation size errors or load errors and must be included in the assessment of hardness as correlation coefficients. The estimated mean absolute square errors for the first two load points are over 10 %. Another critical area is at the end of the composite region, when using a load over 1.5 N. Here, the effect of the substrate hardness becomes significant and the composite hardness increases. The important factor is the depth of the penetration of the top of an indenter. The substrate starts to contribute the measured hardness at the penetration depth 0.07-0.20 times the coating thickness [10].

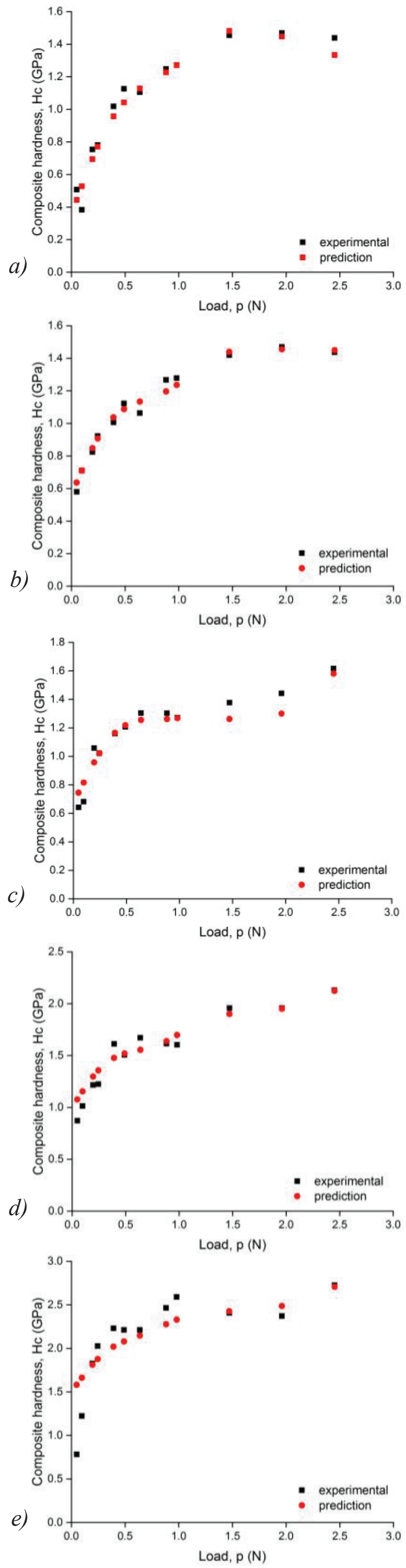

Fig. 2.Comparasion of composite hardness values (experimental and prediction) depending on the applied indentation load for different current density: a) 10 ; b) 33.33; c) 50; d) 66.67; e) 100 mA/cm^2.

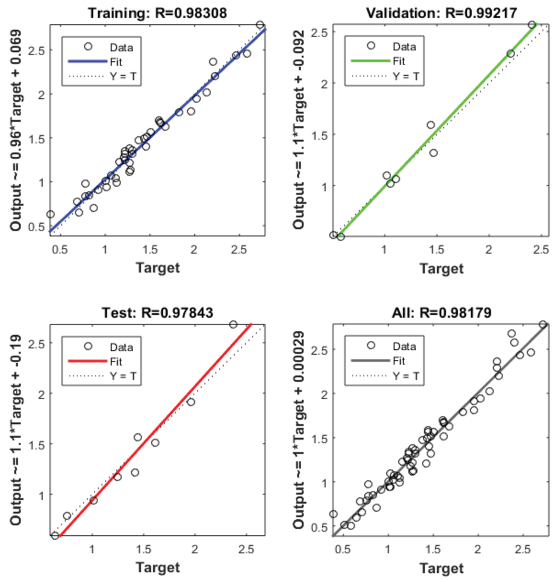

Fig. 3.ANN regression for microhardness modeling.

TABLE I
RESULTS OF THE CRITERIA USED IN PREDICTION COMPOSITE HARDNESS CHANGE

data	samples	MSE	R
training	42	9.489e-3	9.831e-1
validation	9	1.023e-2	9.922e-1
testing	9	2.010e-2	9.784e-1

Predictive ability of the models was evaluated using performance indicators (2) and (3): MSE and R^2 for training, validation and testing data as shown in Table I. Ideal values are MSE=0 and R^2=1. The plot on Fig.3 shows a regression between network outputs and network targets. If the training were perfect, the network outputs and the targets would be equal. The R-values were found as 0.983 for training, 0.922 for validation and 0.978 for testing. With the result above, it is possible to say that the proposed model was well trained and showed an acceptable accuracy in predicting the composite hardness change with variations of current density and applied load.

TABLE II
RESULTS OF THE FILM HARDNESS CHANGE FOR EXPERIMENTAL AND PREDICTION VALUES

sample	experimental		prediction	
j (mA/cm2)	H_f (GPa)	k’	H_f (GPa)	k’
10	0.605	0.133	0.687	0.122
33.33	0.739	0.109	0.852	0.099
50	0.852	0.105	0.921	0.110
66.67	1.056	0.212	0.988	0.204
100	1.591	0.312	1.457	0.386

To estimate the film hardness independently of the substrate Korsunsky model was applied in order to determine absolute hardness of the films. Fitted results are shown in Table II. The increase in the hardness of the film with increasing current density is evident for each sample. The predicted results of the film hardness and experimental values are close.

The next step is predicting the data on which the network was not trained. Three new values of current density were selected (15, 65 and 85 mA/cm^2). The results of prediction film hardness according ANN model are given in Table III. In Fig. 4. prediction of composite hardness for two current densities that are outside the range of experimental measurement are shown.

TABLE III
RESULTS OF THE FILM HARDNESS CHANGE FOR PREDICTION VALUES

sample	Prediction	
j (mA/cm^2)	H_f (GPa)	k’
15	0.724	0.116
65	0.895	0.189
85	0.904	0.403

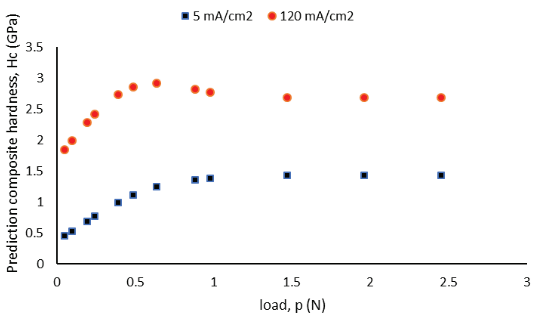

Fig. 4. Predicted composite hardness according to ANN model for current densities of 5 and 120 mA/cm^2.

It can be seen that predicted results follow experimentally established dependencies on current density and load values and are in line with results presented in Fig.2 and Table 2. In the same way, the system's hardness can be predicted for any load point that has not been experimentally performed.

VI. CONCLUSION

Composite systems of electrochemically deposited Cu films on Si (100) substrates were prepared and investigated.

An Artificial Neural Network model was developed and tested for predicting composite hardness of Cu/Si systems using total of 60 experimentally obtained data records. In this article, the focus was on modeling the effects of current density and indentation load on composite and film hardness via ANN predictions.

We have shown excellent consistency and good agreement between ANN predicted results and experimental measurements. Added advantage is that ANN is constantly learning improving with each iteration. Predicting the composite hardness or film hardness over the ANN model is useful when we want to define the properties of a material in advance or to evaluate feasibility of a situation not experimentally performed.

Our future research will focus on more complex ANNs ideally bypassing the need for any analytic approximations in determining the thin film hardness.

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