Response Surface Methodology and Artificial Neural Network-Based Models for Predicting Roughness of Cu coatings

Ivana Mladenović, Jelena Lamovec, Nebojša D. Nikolić, Stevan Andrić, Marko Obradov, Vesna Radojević, Dana Vasiljević-Radović

***Abstract*— Copper coatings are produced on silicon wafer by electrodeposition (ED) in pulsating current (PC) regime. Electrodeposition was performed at various current density amplitudes in the range of 80–140 mA cm^{-2}, frequency in the range of 30–100 Hz and coating thickness in the range of 10–60 µm. The resulting composite systems consist of monolayered copper films electrodeposited from sulfate bath on Si wafers with sputtered layers of Cr/Au. Roughness measurements were performed to evaluate properties of the copper coating surface. The coating roughness (R) was measured using Atomic Force Microscope in contact mode. The software Gwyddion was used for determination an average roughness parameter (R_a). After that (Artificial Neural Network-ANN) model was used to study the relationship between the parameters of electrodeposition process and roughness of copper coatings. The influence of experimental values: amplitude current density, frequency and thickness of coating on the surface roughness will be highlighted. Response surface methodology (RSM) was utilized to improve the correction between R_a and input parameters. Finally, the results of the average roughness (experimental and predicted) were used to estimate the new value of (Ra) of copper for each variation of the input parameters and compared capability of ANN and regression analysis for surface roughness generated under different electrochemical conditions. The coefficient of determination was found 92% for ANN and 93% for regression analysis.**

***Index Terms*—electrodeposition, roughness, AFM, coatings, ANN, RSM.**

Ivana Mladenović is with Institute of Chemistry, Technology and Metallurgy, University of Belgrade, Njegoševa 12, Belgrade, Serbia (e-mail: ivana@nanosys.bg.ac.rs).

Jelena Lamovec is with Institute of Chemistry, Technology and Metallurgy, University of Belgrade, Njegoševa 12, Belgrade, Serbia (e-mail: jejal@nanosys.bg.ac.rs).

Nebojša D. Nikolić is with Institute of Chemistry, Technology and Metallurgy, University of Belgrade, Njegoševa 12, Belgrade, Serbia (e-mail: nnikolic@ihtm.bg.ac.rs).

Stevan Andrić is with Institute of Chemistry, Technology and Metallurgy, University of Belgrade, Njegoševa 12, Belgrade, Serbia (e-mail: stevan@nanosys.bg.ac.rs).

Marko Obradov is with Institute of Chemistry, Technology and Metallurgy, University of Belgrade, Njegoševa 12, Belgrade, Serbia (e-mail: marko.obradov@nanosys.bg.ac.rs).

Vesna Radojević is with the Faculty of Technology and Metallurgy, University of Belgrade, Karnegijeva 4, 11 000 Belgtrade, Serbia (e-mail: vesnar@tmf.bg.ac.rs).

Dana Vasiljević-Radović is with Institute of Chemistry, Technology and Metallurgy, University of Belgrade, Njegoševa 12, Belgrade, Serbia (e-mail: dana@nanosys.bg.ac.rs).

I. INTRODUCTION

Electrodeposition is one of the methods for developing nanocrystalline copper thin coatings with variation surface quality depending on the deposition parameter and the properties of the substrate.

The best benefit of electrodeposition process over other metallization methods such as chemical or physical vapor deposition (CVD or PVD) is speed, simplicity, low cost and the ability to predefine material properties as a function of variable of electrodeposition conditions.

For application in Micro-Electro-Mechanical Systems (MEMS) technologies, coatings must have satisfactory properties such as: good wear and corrosion resistance, compactness, fine microstructure, strong adhesion to the substrate, good mechanical durability and ductility and low roughness [1-5]. The surface roughness is an important indicator of the quality of electrodeposited metallic coatings. Also, the mechanical properties, such as microhardness [4], creep life and fatigue behavior [6] and electrical properties like conductivity [7] depend on the surface roughness of the coatings.

Measuring the surface roughness of a coating by scanning microscopy, such as AFM is a time-consuming process as is the processing of each individual image, but is a starting point for collecting of the roughness data, necessary to monitor the quality of the coatings.

By the use of the developed software package, we are able to design an experiment, predictable behavior of materials with variation of process parameters and generate a model that best describes the system we designed.

Artificial neural network (ANN) is powerful tools for modeling, especially when the data are randomly distributed and when the connection between the input parameters is unknown [8-10]. Response Surface Methodology (RSM) represents a collection of mathematical and statistical techniques that are useful for the modeling, analysis data sets and find the mathematical correlation between the input variables and the output response [11-12].

Both methods (ANN and RMS) can be used to predict the values of output variables (mainly the measured values) as a function of input variables (process parameters) depending on the type of system and the quality of prediction we want to achieve [13].

II. EXPERIMENTATION

2.1. Materials and experimental conditions

Electrodeposition of copper was performed from sulfate bath, composition: 240 g /L $CuSO_4 \cdot 5 H_2O$, 60 g /L H_2SO_4. The deposition was performed in an open-type cell at the room temperature by the regime of pulsating current (PC).

The parameters of the PC regime used in the processes of electrodeposition are given in Table 1. The current density amplitude (j_A in mA cm^{-2}), frequency (f in Hz) and coating thickness (δ in µm) are varied parameters. Deposition pulse time (t_c in ms) was kept constant at 5 ms.

TABLE I: DEPOSITION PARAMETERS.

No.	j_A/ mAcm^{-2}	f/ Hz	δ/ µm
1	80	100	40
2	100	100	40
3	120	100	40
4	140	100	40
5	100	30	40
6	100	50	40
7	100	80	40
8	100	100	10
9	100	100	20
10	100	100	60

For these experiments, substrate of Si wafers (2'', (111) orientation) was chosen and prepared to serve as a cathode. The wafer was cut in parts (1 × 1) cm^2 surface area, standard cleaning and drying procedures. The plating base on the silicon wafers were sputtered layers of 30 nm Cr and 100 nm Au, using Perkin Elmer 2400 sputtering system. High purity copper was used as an anode.

2.2. Roughness measurement of copper coatings

The surface topography and roughness of the Cu coatings were examined using atomic force microscope (AFM, TM Microscopes-Vecco) in the contact mode. The values of the arithmetic average of the absolute roughness parameters (R_a) of the surface height deviation, were measured from the mean image data plane, using software Gwyddion. The values of R_a roughness parameter, calculated as average from three independent measurements at different locations of one sample of copper surface obtained by the PC regime with variation of electrodeposition conditions. For three different scan areas (2500, 4900 and 8100) µm^2, two measurements were made, and average roughness parameter was calculated.

The average roughness can be calculated as:

$$R_a = \frac{1}{N_x \cdot N_y} \sum_{i=1}^{N_x} \sum_{j=1}^{N_y} \left| z(i,j) - z_{mean} \right| \quad (1)$$

where N_x and N_y are the number of scanning points on the x-axis and y-axis; $z(i,j)$ is the height of the (i,j) measuring point, z_{mean} is the mean high of all measuring points [14].

III. RESULTS AND DISCUSSION

The AFM surface areas of Cu coatings produced at different thickness (10 and 60 µm) are shown in Fig. 1. The values of R_a parameter obtained for various thickness of coatings for three scan area surface area are shown in Table II.

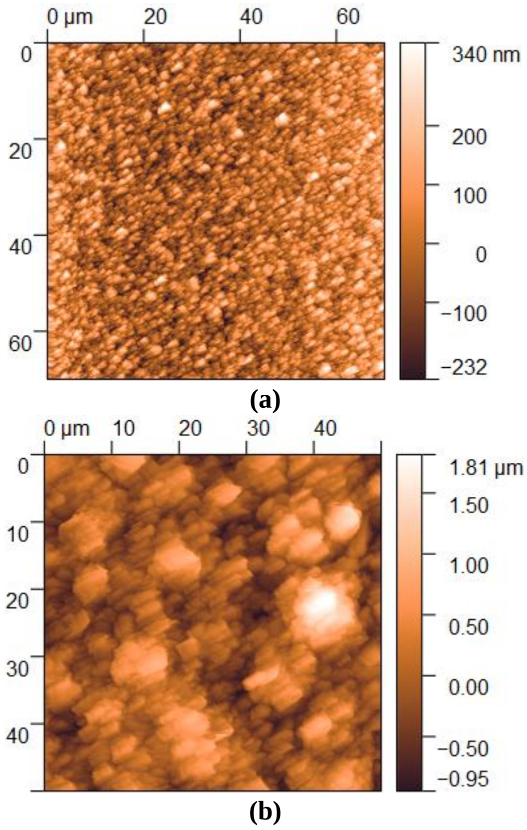

Fig. 1. The AFM images of Cu coatings obtained by the PC regimes: a) δ = 10 µm, b) δ = 60 µm. j_A= 100 mA cm^{-2}, f = 100 Hz, t_c = 5 ms. Scan area was 2500 µm^2.

TABLE II
Roughness variation with coating thickness and constant frequency and amplitude current density; j_A =100 mA cm^{-2} and f = 100 Hz.

δ/ µm	*Scan area*/ µm × µm	R_a/ nm	δ/ µm	*Scan area*/ µm × µm	R_a/ nm
10	50 × 50	66.54	20	50 × 50	114.7
10	50 × 50	65.12	20	50 × 50	121.9
10	70 × 70	65.81	20	70 × 70	121.2
10	70 × 70	60.82	20	70 × 70	120.4
10	90 × 90	64.71	20	90 × 90	125.9
10	90 × 90	74.99	20	90 × 90	119.7
40	50 ×50	280.8	60	50 × 50	299.9
40	50 × 50	277.9	60	50 × 50	325
40	70 × 70	249.3	60	70 × 70	309.4
40	70 × 70	246.6	60	70 × 70	309.2
40	90 × 90	247.1	60	90 × 90	297.5
40	90 × 90	251.6	60	90 × 90	292.9

3.1 Artificial Neural Network (ANN)

MATLAB R2017b and *nntool* box was chosen and applied on roughness experimental data for training, testing, validation and prediction of new values of R_a. In the present work, feedforward back propagation network was utilized. The best network structure for this set data was 4-10-1, 4 corresponding to the input layer neurons, 10 to hidden layer neurons and 1 to output layer neurons. The numerical technique for optimization was Levenberg-Marquardt (LM).

The plot on Fig. 2 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 0.99608 for training (Fig. 2a), 0.99636 for validation (Fig. 2b), 0.99176 for testing (Fig. 2c) and 0.99553 for all data set (Fig. 2d).

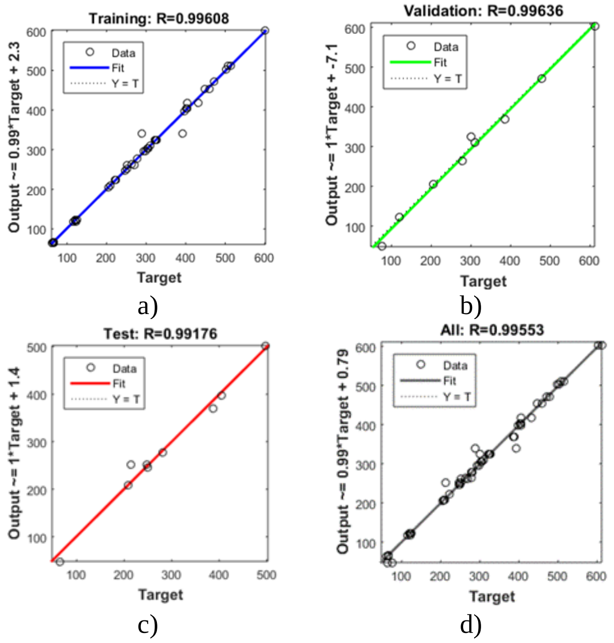

Fig. 2. The ANN regression for roughness parameter modeling, a) training data (70% of data set), validation data (15%) and testing (15%).

Based on the values of the regression coefficient values (*R*) and values of the mean relative error (MRE) ANN predictive model is satisfactory and adequate. Fig. 3 shows the experimental and the corresponding ANN predicted values as well as the relative error for each data. The mean absolute percentage error (MAPE) for the ANN model was 7.84 %, calculated from the data in Fig. 3.

The maximal error was detected for sample with lower scanned area, which was expected due to the sensitivity of the measurement method. For a realistic estimation of the roughness of the samples it is necessary to make measurements at different locations of the sample and at different scanned surfaces.

3.1 Response Surface Methodology (RSM)

Design-Expert 12 software and response surface central-composite-design was employed for experimental design. A small data set was used, for a constant scan area value of 2500 μm^2 for all samples. The factors and factor levels are shown in Table III.

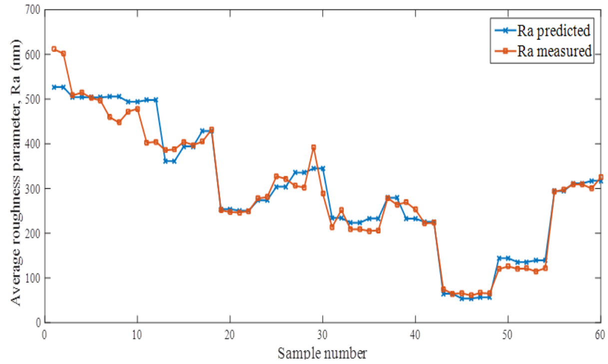

Fig. 3. Experimental versus predicted values of surface roughness using ANN.

TABLE III The factors levels.

Factors	Levels			
Amplitude of current density	80	100	120	140
Frequency	30	50	80	100
Coating thickness	10	20	40	60

The analysis of variance (ANOVA) was utilized in testing the roughness data. The linear-model generated by the following equation:

$$Y_u = \beta_0 + \sum_{i=1}^{n} \beta_i x_i + e \qquad (2)$$

where Y_u is the required response, β_0, β_1... β_i are the regression coefficients, *x* is the independent variables and *e* is the error.

The final equation in terms of coded factors is given as:

$$R_a = 506.9598 - 1.11778\, j_A - 3.63598\, f + 5.39583\, \delta \qquad (3)$$

The equation (3) can be used to make predictions about the response for given levels of each factor. By default, the high levels of the factors are coded as +1 and the low levels are coded as -1. The coded equation is useful for identifying the relative impact of the factors by comparing the factor coefficients. We can see that thickness is dominated factor on influence to the roughness copper coatings. Fig. 4 shows the residuals against run test and data points are randomly distributed.

Fig. 5 shows the predicted response vs. the actual value points are distributed along a 45° line; this means that the organized model is adequate and satisfactory.

Fig. 6 is a 3D view response surface curve for surface roughness. The best roughness value can be obtained at 100 Hz with current density at 100 mA cm^{-2}, for small coating thickness.

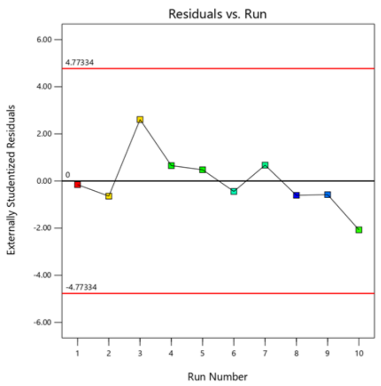

Fig. 4. Residuals against run tests for surface roughness parameter.

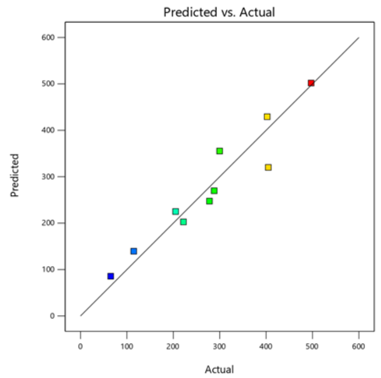

Fig. 5. Predicted response *vs*. the actual values for surface roughness parameter for copper coatings.

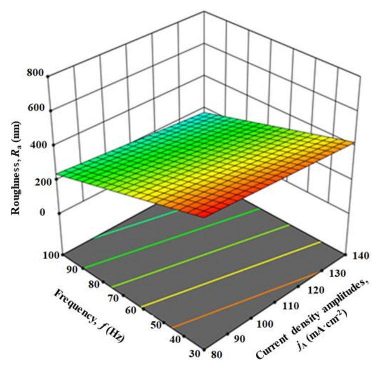

Fig. 6. 3D response surface curves for the surface roughness. A coating thickness of 35 μm was selected for prediction.

IV. Conclusion

The conditions suitable for formation of compact and uniform Cu coatings of a satisfactory roughness by the regime of pulsating current (PC) were defined. The Cu coatings were obtained with variation of an amplitude of current density, frequency and coating thickness. The minimal roughness showed the Cu coatings obtained at 100 Hz with the current density amplitude of 100 mA cm^{-2} and coating thickness at 10 μm. From the results obtained by application of two different models follows:

-The coating thickness has strong influence on micro roughness;

-The current density also strongly affected the coating roughness, but less than frequency.

-Both RSM and ANN show good agreement with experimental results.

-Both methods can be effectively used to predict coating roughness.

Acknowledgment

This work was financially supported by the Ministry of Education, Science and Technological Development of the Republic of Serbia (Grants No. 451-03-68/2020-14/200026 and No. 451-03-68/2020-14/200135).

References

[1] A. Maciossek, B. Lochel, H. J. Quenzer, B. Wagner, S. Schulze, J. Noetzel, "Galvanoplating and sacrificial layers for surface micromachining", *Microelectron. Eng.* , vol. *27*, no.*1*, pp. *503-508*, Feb., 1995.

[2] Z. Zhao, L. Du, Z. Xu, L. Shao, "Effects of ultrasonic agitation on adhesion strength of micro electroforming Ni layer on Cu substrate," *Ultrason. Sonochem.*, vol. *29*, no./, pp. *1-10*, Mar., 2016.

[3] S. Banthia, S. Sengupta, S. Das, K. Das, "Cu, Cu-SiC functionally graded coating for protection against corrosion and wear", *Surf. Coat. Technol.*, vol. *374*, no. , pp. *833-844*, Sept., 2019.

[4] I. Mladenović , J. Lamovec, V. Jović, M. Obradov, D. Vasiljević-Radović, N. D. Nikolić, V. Radojević, "Mechanical characterization of copper coatings electrodeposited onto different substrates with and without ultrasound assistance", *J. Serb. Chem. Soc.* , vol. *84*, no. *7*, pp.*729-741*, Mar., 2019.

[5] J.B. Marro, T. Darroudi, C.A. Okorod, Y.S. Obeng, K.C. Richardson, "The influence of pulse plating frequency and duty cycle on the microstructure and stress state of electroplated copper films", *Thin Solid Films*, vol. *621*, no. , pp. *91–97*, Jan., 2017.

[6] B. Wilshire, A.J. Battenbough, "Creep and creep fracture of polycrystalline copper", *Mater. Sci. Eng. A*, vol. *443*, no./, pp. *156-166*, Aug., 2007.

[7] A. Javidjam, M. H. Hekmatshoar, L. Hedayatifar, S. N. K. Abad, "Effect of surface roughness on electrical conductivity and hardness of silver plated copper", *Mater. Res. Express*, vol. *6*, no. *3*, pp. *036407*, Dec., 2019.

[8] I. Mladenović, J. Lamovec, V.Jović, M. Obradov, K. Radulović, D. Vasiljević-Radović, V. Radojević, "Artificial Neural Network for Composite hardness Modeling of Cu/Si Systems Fabricated Using Various Electrodeposition Parameters", Proc. of the 31th International Conference on Microelectronics MIEL, Niš, Serbia, pp. 133-136, 16.-18. Sept., 2019.

[9] D. M. Habashy, H. S. Mohamed, E. F. M. El-Zaidia, "A simulated neural system (ANNs) for micro-hardness of nano-crystalline titanium dioxide", *Physica B: Condensed Matter.*, vol. *556*, pp.*183-189*, Mar., 2019.

[10] M.S. Ozerdem, S. Kolukisa, "Artificial neural network approach to predict the mechanical properties of Cu-Sn-Pb-Zn-Ni cast alloys", *Materials and design*, vol. *30*, no. *3*, pp.*764-769*, Mar., 2009.

[11] G. Orhan, G. Hapci, O. Keles, "Application of Response Surface Methodology (RSM) to Evaluate the Influence of Deposition Parameters on the Electrolytic Cu-Zn Alloy Powder", *Int. J. Electrochem. Sci.* , vol. *6*, no. , pp. *3966-3981*, Sept. , 2011.

[12] V. Ponraj, A.Azhagurajn, S. C. Vettivel, S. Shajan, P. Y. Nabhiraj, A. Haiterlenin, "Modeling and Optimization of the Effect of Sintering Parameters on the Hardness of Copper/Graphene Nanosheet Composites by Response Surface Methodology" , *Met. Sci. Heat Tret.*, vol. *60*, no. *9*, pp. *611-615*, Mar., 2019.

[13] A. A. Elsadek, A. M. Gaafer, S.S. Mohamed, "Surface roughness prediction in hard-turning with ANN and RSM", Journal of the Egyptian society of Tribology, vol. *17*, no. *2*, pp. *13-22*, Apr., 2020.

[14] Y. Li, J. Yang, Z. Pan, W. Tong, "Nanoscale pore structure and mechanical property analysis of coal: An insight combining AFM and SEM images", *Fuel*, vol. 260, no. , pp. *116352*, Jan., 2020.