Available online at www.sciencedirect.com

ScienceDirect

Ceramics International 40 (2014) 5425–5430

CERAMICS INTERNATIONAL

www.elsevier.com/locate/ceramint

Application of artificial neural networks to predict corrosion behavior of Ni–SiC composite coatings deposited by ultrasonic electrodeposition

Youjun Xu[a,*], Yongyong Zhu[b], Guorong Xiao[c], Chunyang Ma[d]

[a]College of Computer Science and Information Technology, Daqing Normal University, Daqing 163712, PR China
[b]Department of Economics and Business Administration, Chongqing University of Education, Chongqing 400067, PR China
[c]Department of Computer Science and Technology, GuangDong University of Finance, Guangzhou 510521, PR China
[d]School of Mechanical Science and Engineering, Northeast Petroleum University, Daqing 163318, PR China

Received 5 October 2013; received in revised form 23 October 2013; accepted 25 October 2013
Available online 13 November 2013

Abstract

A feed-forward, multilayer perceptron artificial neural network (ANN) model with eight hidden layers and 12 neurons was used to predict the corrosion behavior of Ni–SiC composite coatings deposited by ultrasonic electrodeposition. The effect of process parameters, namely, ultrasonic power, SiC particle concentration, and current density, on the weight losses of Ni–SiC composite coatings was investigated. The grain sizes of Ni and SiC were determined by using X-ray diffraction (XRD) and scanning probe microscopy (SPM). Results indicate that ultrasonic power, SiC particle concentration, and current density have significant effects on the weight losses of Ni–SiC composite coatings. The ANN model, which has a mean square error of approximately 3.35%, can effectively predict the corrosion behavior of Ni–SiC composite coatings. The following optimum conditions for depositing Ni–SiC composite coatings were determined on the basis of the lowest weight loss of Ni–SiC deposits: ultrasonic power of 250 W, SiC particle concentration of 8 g/l, and current density of 4 A/dm^2. XRD and SPM results demonstrate that the average grain sizes of Ni and SiC in the Ni–SiC composite coating are 90 and 70 nm, respectively.

Keywords: A. Artificial neural networks; B. Prediction; C. Ni–SiC composite coating

1. Introduction

Electrodeposition is a well known technique for the preparation of excellent performance coatings due to their enhanced microhardness, better wear and corrosion resistance when compared to pure metal or alloy [1–4]. Recent literatures on the electrodeposition of composite coatings are studied. Borkar et al. [5] demonstrated that pulse electrodeposits featuring codeposited CNTs exhibit finer mean matrix grain size, with the grain refinement increasing with increasing volume fraction of CNTs in the electrodeposition. Parida et al. [6] reported that the Ni–TiO_2 composite coating on steel substrates was directly prepared by electrodeposition from a bath containing dispersion of TiO_2 power in Watt's bath. They found that TiO_2 particles of less than 100 nm size were homogeneously co-deposited with nickel on steel substrate, and microhardness values were increased after incorporation of TiO_2 compared to pure nickel deposition. Xia et al. [7] obtained the electroplating parameters for preparation of Ni–TiN composite coating with high micro-hardness and excellent corrosion resistance by direct current (DC), pulse current (PC) and ultrasonic pulse current (UPC) deposition methods. It showed that the coating prepared by UPC deposition exhibits the best corrosion resistance, whereas the coating fabricated by DC deposition suffers the most serious damage. It is believed that the introduction of superfine particles, such as AlN, CNTs TiO_2, and SiC, into the coatings often result in grain refinement or change in microstructure of the matrix [8–11].

Ultrasonic electrodeposition, a method involving electronic agitation to suspend particles in electrolyte, is more effective compared with mechanical stirring. However, ultrasonic electrodeposition is influenced by parameters, such as current density, ultrasonic power, particle concentration, on-duty ratio,

*Corresponding author. Tel./fax: +86 459 6505777.
E-mail address: youjunxu327@163.com (Y. Xu).

0272-8842/$ - see front matter © 2013 Elsevier Ltd and Techna Group S.r.l. All rights reserved.
http://dx.doi.org/10.1016/j.ceramint.2013.10.125

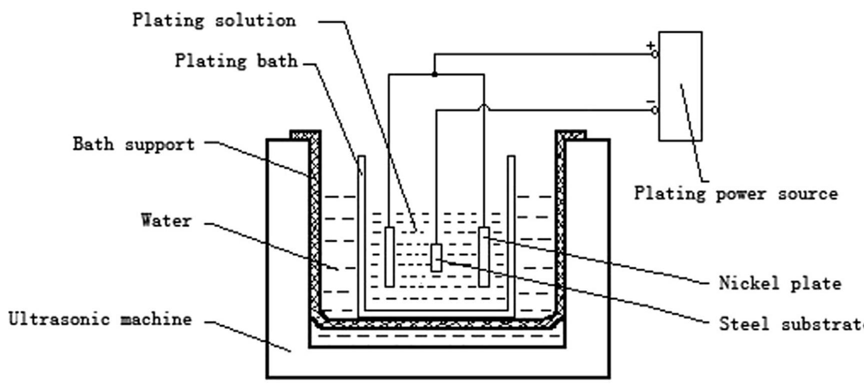

Fig. 1. Schematic diagram of the ultrasonic electrodeposition cell.

and bath temperature. A schematic diagram of a basic ultrasonic electrodeposition cell is shown in Fig. 1. Exploiting the net surface electrostatic characteristics of stable non-agglomerated SiC particles in colloidal suspension is an effective method in infiltrating densely packed steel. In an aqueous suspension within an electric field, SiC particle movement is governed by electric pulse parameters, electrolyte pH, ultrasonic power, additive content, and so on.

Many investigations have been made on electrodeposition or ultrasonic electrodeposition of metals. However, few reports exist on the application of artificial neural networks (ANNs) in predicting the corrosion behavior of Ni–SiC composite coatings. In the present study, Ni–SiC composite coatings were synthesized by means of ultrasonic electrodeposition method. Experimental dataset for the preparation of Ni–SiC composite coatings were investigated. The corrosion behavior of Ni–SiC composite coatings was modeled by using ANNs.

2. Experimental

Ni–SiC composite coatings with thickness of $\sim 100\ \mu m$ were deposited on the $30\ mm \times 20\ mm \times 5\ mm$ mild steel substrates by ultrasonic electrodeposition. And the thickness of the coatings was measured by an ultrasonic thickness detector (ELEKTROPHYSIK, MiniTest600B-FN). The mild steel substrates were used as the cathodes. Prior to deposition, the substrates were mechanically polished to a $\sim 0.15\ \mu m$ surface finish, sequentially cleaned to remove surface contamination, activated for 10 s in a mixed acidic bath, rinsed with distilled water and ethyl alcohol. A similar dimension of pure nickel (99.99%) plate was used as the anode. In order to obtain electrodeposited Ni–SiC composite coatings, the composition of the electrolyte was as follows: 300 g/l nickel sulfate, 45 g/l nickel chloride, 25 g/l boric acid and 2–10 g/l SiC particles. The temperature was kept at 50 °C at pH 4.5, adjusted using ammonium hydroxide or dilutes sulfuric acid. During electrodeposition, the SiC particles in the range of ~ 50 nm were introduced into the electrolyte in various ratios. The plating parameters for ultrasonic electrodepositing Ni–SiC composite coatings are shown in Table 1.

Corrosion tests were carried out on the Ni–SiC composite coatings by immersing samples in 5 wt% NaCl solution for 200 h at 30 °C, then rinsed with distilled water, and finally dried in a drying oven (CIXI, FY-DR-1). The weight loss was measured on an electronic analytical balance (SARTORIUS, BS210S) with an accuracy of 0.01 mg. The surface morphology of the coatings was observed by SPM (Digital Instruments, Nanoscope IIIa). To determine the phase structu re of Ni–SiC composite coatings, XRD analysis was performed on a Rigaku D/Max-2400 instrument using Cu Kα radiation ($\lambda = 0.15418$ nm). The operating target voltage was 40 kV and the tube current was 100 mA. Using the Scherrer equation, the average grain diameter can be calculated as follows:

$$D = \frac{180K\lambda}{\pi\sqrt{\beta^2 - \omega}\ \cos\ \theta} \tag{1}$$

where K is the figure factor of the grains ($K = 0.89$), λ is the wavelength, β is the width of the diffraction peak at half height, ω is the standard Full Width at Half Maximum (FWHM) and θ is the Bragg angle.

Table 1
Plating parameters for preparing Ni–SiC composite coatings.

Parameters	Series I	Series II	Series III
Ultrasonic power (W)	100		
	150		
	200		
	250	250	250
	300		
	400		
	500		
SiC particle concentration (g/l)		2	
		4	
	8	6	8
		8	
		10	
Current density (A/dm^2)			2
			3
	4	4	4
			5
			6
Pulsed frequency (Hz)	100	100	100
Electroplating time (min)	90	90	90

3. Model setup

ANNs are composed of simple artificial nodes that can mimic biological neural networks when connected. The simple neural network was first introduced by McCulloch and Pitts (1943). Currently, ANN tends to refer mostly to neural network models employed in statistics, cognitive psychology, and artificial intelligence. Fig. 2 presents the framework of the ANN model, which comprises input, hidden, and output layers.

This study used feed-forward, multilayer perceptron, which was trained with back propagation algorithm. Ultrasonic power (U_p), SiC particle concentration (C_{sic}), and current density (C_d) were used as inputs, whereas weight loss was considered as the output of the neural network model. Inputs and outputs were normalized within the range of 0–1 [12,13]. The output y_i produced by neuron i in layer L is described as follows:

$$y_i = f\left(\sum_{j=1}^{n} W_{ij} + b\right) \tag{2}$$

where f is the activation function, n is the number of elements in the layer L-1, and b is the offset or bias where the activation function shifts along the basic axis; W_{ij} is the weight associated with the connection between neuron i in layer L and neuron j in layer L-1, which has an output of w_i.

The mean square error (MSE) is expressed by the following relationship:

$$MSE = \frac{1}{NT}\sum_{m=1}^{T}\sum_{n=1}^{N}[d_i(m) - y_i(m)]^2 \tag{3}$$

where N is the number of outputs, T is the number of training sets, d_i is the desired output, and y_i is the network output.

4. Results and discussion

4.1. Effect of ultrasonic powers on the weight losses of Ni–SiC composite coatings

Fig. 3 shows the weight loss variation of Ni–SiC composite coatings as a function of ultrasonic power. The weight loss of Ni–SiC coatings decreased as the ultrasonic power increased to 250 W. However, this weight loss increased with ultrasonic power higher than 250 W. This phenomenon can be attributed by the homogeneous dispersion of SiC particles in the electrolyte, which is triggered by moderate ultrasonication. On the contrary, higher ultrasonic power dislodges SiC particles from the substrate into the electrolyte, leading to lower SiC particle content in the coatings. Such a behavior has been experimentally observed for some crystalline deposits [14,15].

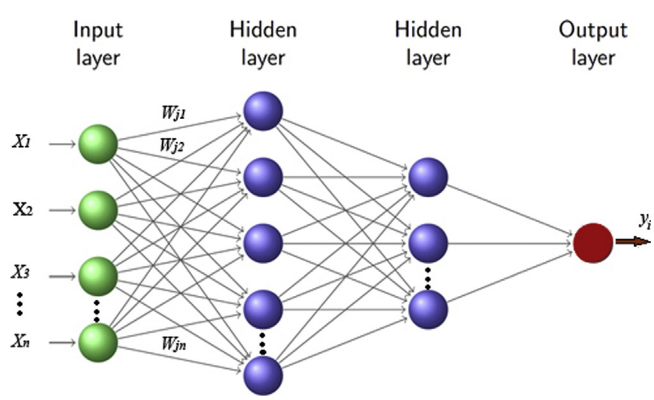

Fig. 2. A schematic description of artifical neural network configuration.

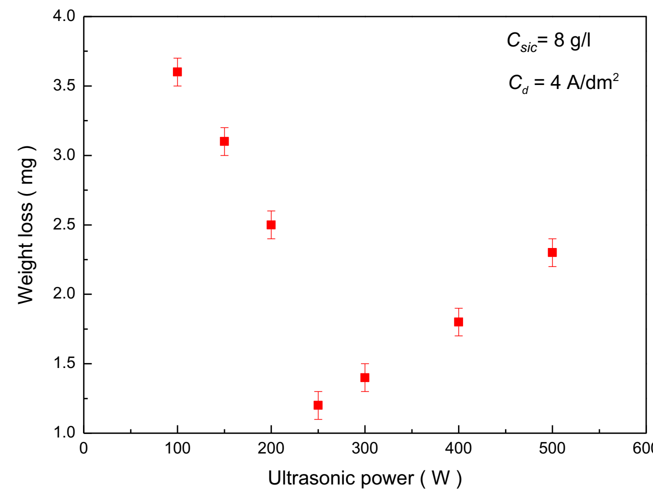

Fig. 3. Effect of ultrasonic powers on the weight losses of Ni–SiC composite coatings.

4.2. Effect of SiC particle concentrations on the weight losses of Ni–SiC composite coatings

Fig. 4 shows the weight loss variation of Ni–SiC composite coatings as a function of SiC particle concentrations. As shown in Fig. 4, the weight loss of the composite coatings decreases greatly when the SiC particle concentration is increased and then leveled off at 8 g/l of SiC particles. However, the weight loss of the composite coatings increases slightly when SiC particle concentration increases from 8 g/l to 10 g/l. A similar effect was reported by Corni et al. [16] and Yuan et al. [17]. This phenomenon can be attributed to the combined effects of retarding surface diffusion and blocking the crystalline growth, hydrogen evolution, and changes in overpotential.

4.3. Effect of current densities on the weight losses of Ni–SiC composite coatings

Fig. 5 displays the effect of current densities on weight loss variations of Ni–SiC composite coatings. Current density has an obvious effect on the weight loss of deposits. The weight loss of composite coatings decreases when the current density is increased to 4 A/dm^2. However, further increase in current density has no significant effect on weight change. Increasing the current density results in high overpotential, which leads to increased nucleation rate. Increasing the current density also leads to Ni–SiC composite coatings with compact and exiguous surface morphologies, as exhibited in the general patterns presented by Vaezi et al. [18] and Benea et al. [19]. Moreover,

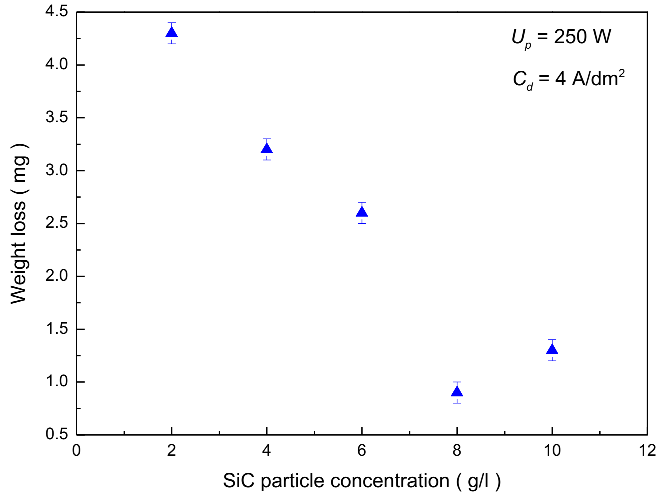

Fig. 4. Effect of SiC particle concentrations on the weight losses of Ni–SiC composite coatings.

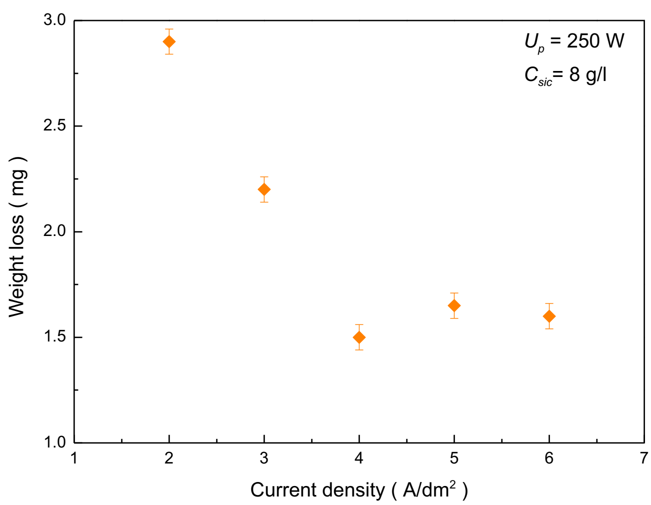

Fig. 5. Effect of current densities on the weight losses of Ni–SiC composite coatings.

moderate ultrasonication also results in the homogeneous dispersion of SiC particles in the coatings. The weight loss of the coatings slightly increases when the current density increases from 4 A/dm^2 to 6 A/dm^2.

4.4. Modeling results

4.4.1. Number of hidden layers and neurons

A major problem in designing neural networks is establishing the required number of hidden layers and neurons. Using an excessive number of hidden neurons will cause overfitting, which denotes that neural networks have overestimated the complexity of the target problem. This overestimation greatly degrades generalization capability, which leads to significant deviation in predictions. Thus, determining the proper number of hidden neurons that will not cause overfitting is critical in function approximation using ANN.

The MSE demonstrates the accuracy of prediction. Fig. 6 illustrates the MSE for various hidden layers and shows that the ANN with eight hidden layers and 12 neurons yields the smallest error. Therefore, the ANN model with these properties was used to predict the corrosion behavior of Ni–SiC composite coatings.

4.4.2. Model predictions

Fig. 7 presents a comparison between the experimental and predicted weight losses of Ni–SiC composite coatings at different ultrasonic powers, SiC particle concentrations, and current densities. The figure presents a linear relationship between the experimental results and predictions from the ANN model, which determines the reliability of the model.

Fig. 8 illustrates the predicted values of weight losses in comparison with experimental values as a function of ultrasonic power, SiC particle concentration, and current density.

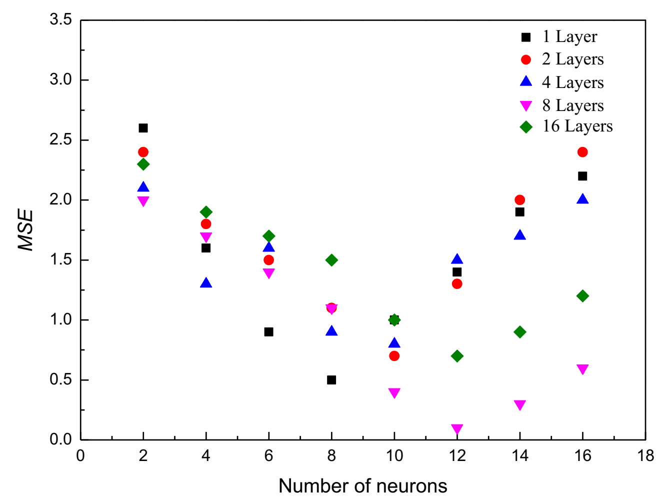

Fig. 6. *MSE* for different hidden layers and neuron numbers.

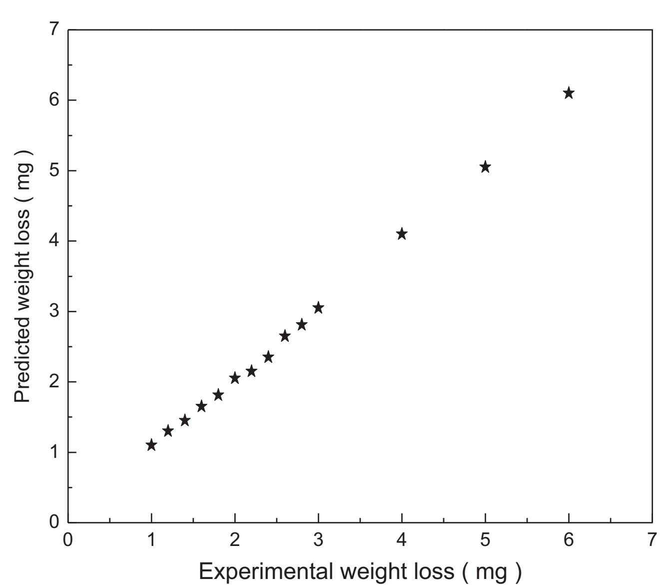

Fig. 7. Relationship between the experimental and predicted weight losses of Ni–SiC composite coatings.

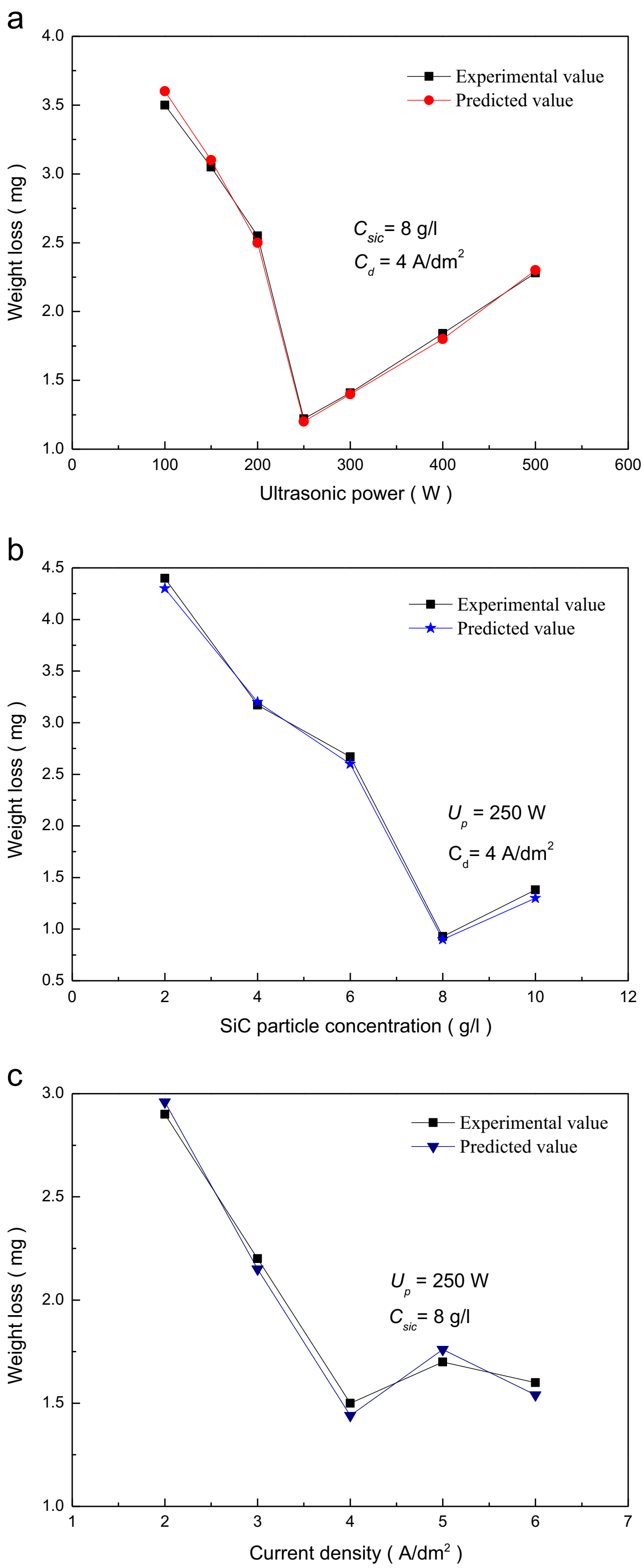

Fig. 8. Comparison between the experimental and predicted weight losses of Ni–SiC coatings deposited at different (a) ultrasonic powers, (b) SiC particles concentrations and (c) current densities.

The predicted values tally with the experimental values, and the smallest weight loss of Ni–SiC deposits is obtained at the ultrasonic power of 250 W, SiC particle concentration of 8 g/l, and current density of 4 A/dm^2. By using Eq. (3) and the predicted data, we have calculated an *MSE* of 3.35% for this ANN model. Thus, the ANN model can be used in predicting the corrosion behavior of Ni–SiC composite coatings.

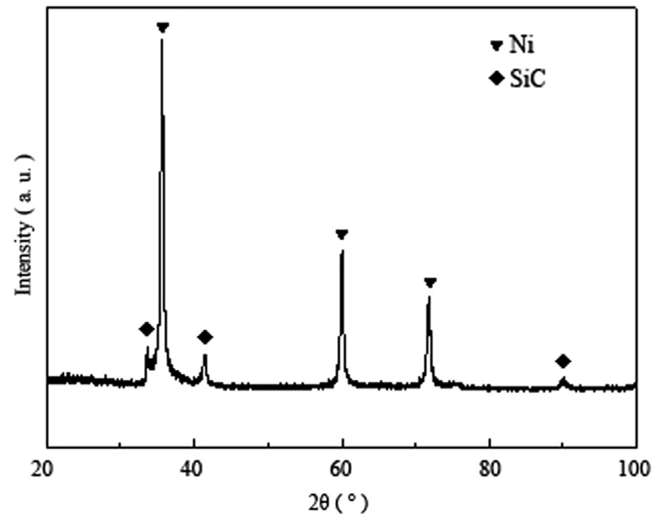

Fig. 9. XRD pattern of the Ni–SiC composite coating.

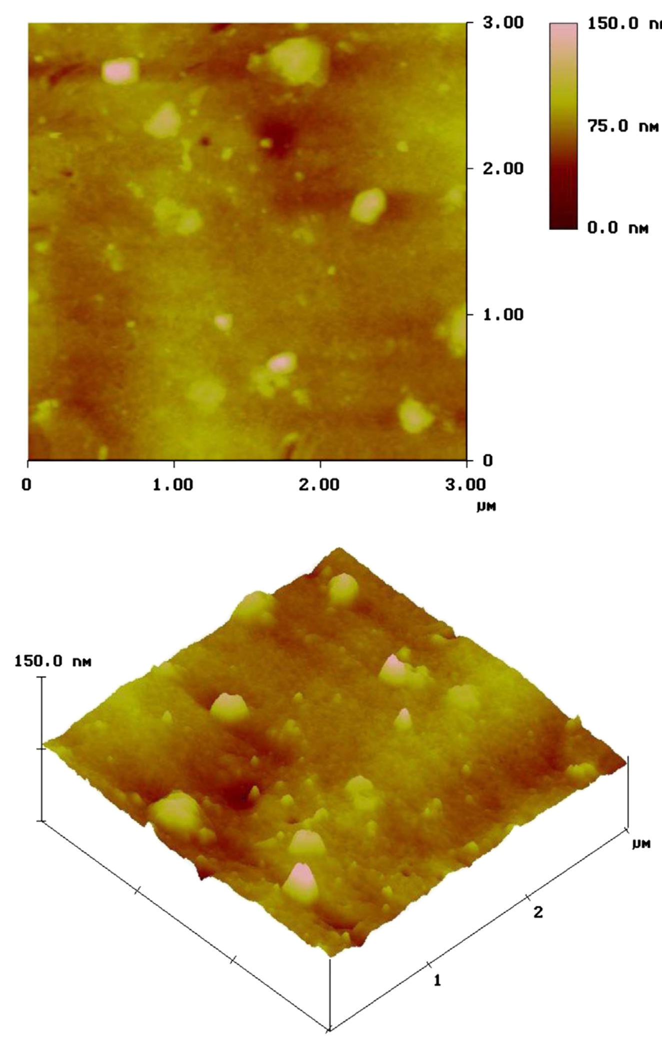

Fig. 10. SPM images of the Ni–SiC composite coating.

4.5. Microstructural analysis

XRD was applied on the Ni–SiC composite coatings, which were obtained at ultrasonic power of 250 W, SiC particle concentration of 8 g/l, and current density of 4 A/dm^2, to confirm further the presence of SiC particles in the coatings (Fig. 9). Scans were recorded for the range $2\theta = 20–100^\circ$ with a scan step of 0.02°. The figure shows that the composite coating consists of Ni phase and SiC phase. For Ni, the diffraction peaks at 44.82°, 52.21°, and 76.77° correspond to (111), (200), and (220). For SiC, the diffraction peaks at 33.68°, 41.38°, and 91.94° correspond to (111), (200), and (220), respectively. According to the XRD data, the average grain size of Ni and SiC can be calculated using Eq. (1). The XRD results demonstrate that the average grain sizes of Ni and SiC in the composite coating prepared by ultrasonic electrodeposition are 89.4 and 65.7 nm, respectively.

To determine the microstructure of the coatings and the size of the grains, the coating surface was observed by SPM. Two- and three-dimensional coating images are shown in Fig. 10. It is evident that the coating contained SiC particles of approximately 70 nm in diameter, larger than the size calculated from XRD analysis. The average diameter of Ni grains is approximately 90 nm.

Taking all the calculations into consideration, the average diameter of SiC particles is 70 nm and the average diameter of Ni grains is 90 nm.

5. Conclusions

(1) The weight loss of Ni–SiC coatings decreases as the ultrasonic power increases to 250 W. However, the weight loss of Ni–SiC coatings increases with further increase in ultrasonic power. The weight loss of the composite coatings decreases greatly when the SiC particle concentration increases, but levels off at 8 g/l of SiC particles. However, weight loss increases slightly when the SiC particle concentration increases from 8 g/l to 10 g/l. The weight loss of the coatings decreases when the current density increases up to 4 A/dm^2, whereas further increase in current density has no significant effect on weight change.

(2) By analyzing the weight loss of Ni–SiC deposits, we have determined that the optimum condition for depositing Ni–SiC composite coatings is at an ultrasonic power of 250 W, SiC particle concentration of 8 g/l, and current density of 4 A/dm^2. The XRD and SPM results demonstrate that the average grain sizes of Ni and SiC in the Ni–SiC composite coating are 90 and 70 nm, respectively.

(3) The ANN model, which has an *MSE* of approximately 3.35%, is an applicable method in predicting the corrosion behavior of Ni–SiC composite coatings.

Acknowledgments

This work has been supported by the Natural Science Foundation of China (Granted no. 51374072), Scientific Research Fund of Heilongjiang Provincial Education Department (Granted no. 12533002).

References

[1] S. Xie, X.L. Tong, G.Q. Jin, Y. Qin, X.Y. Guo, CNT-Ni/SiC hierarchical nanostructures: preparation and their application in electrocatalytic oxidation of methanol, J. Mater. Chem. A 1 (2013) 2104–2109.

[2] Y. Wang, H.B. Xu, Preparation and numerical simulation of Ni–SiC composite coatings deposited by electrode position, Res. J. Appl. Sci., Eng. Technol. 5 (24) (2013) 5602–5607.

[3] M.J. Jiao, X.X. Wang, Electrolytic deposition of magnesium-substituted hydroxyapatite crystals on titanium substrate, Mater. Lett. 63 (2009) 2286–2290.

[4] E.A. Pavlatou, M. Stroumbouli, P. Gyftou, N. Spyrellis, Hardening effect induced by incorporation of SiC particles in nickel electrodeposits, J. Appl. Electrochem. 36 (2006) 385–394.

[5] T. Borkar, S. Harimkar, Microstructure and wear behaviour of pulse electrodeposited Ni-CNT composite coatings, Surf. Eng. 27 (7) (2011) 524–530.

[6] G. Parida, D. Chaira, M. Chopkar, A. Basu, Synthesis and characterization of Ni–TiO_2 composite coatings by electro-co-deposition, Surf. Coat. Technol. 205 (2011) 4871–4879.

[7] F.F. Xia, C. Liu, C.H. Ma, D.Q. Chu, L. Miao, Preparation and corrosion behavior of electrodeposited Ni–TiN composite coatings, Int. J. Refract. Met. Hard Mater. 35 (2012) 295–299.

[8] F.F. Xia, H.B. Xu, C. Liu, J.W. Wang, J.J. Ding, C.H. Ma, Microstructures of Ni–AlN composite coatings prepared by pulse electrodeposition technology, Appl. Surf. Sci. 271 (2013) 7–11.

[9] L. Chen, L. Wang, Z. Zeng, T. Xu, Influence of pulse frequency on the microstructure and wear resistance of electrodeposited Ni–Al_2O_3 composite coatings, Surf. Coat. Technol. 201 (2006) 562–599.

[10] S. Xie, X.L. Tong, G.Q. Jin, Y. Qin, X.Y. Guo, CNT-Ni/SiC hierarchical nanostructures: preparation and their application in electrocatalytic oxidation of methanol, J. Mater. Chem. A 1 (2013) 2104–2109.

[11] E.A. Pavlatou, M. Stroumbouli, P. Gyftou, N. Spyrellis, Hardening effect induced by incorporation of SiC particles in nickel electrodeposits, J. Appl. Electrochem. 36 (2006) 385–394.

[12] S.H. Mousavi Anijdan, A. Bahrami, H.R. Madaah Hosseini, A. Shafyei, Using genetic algorithm and artificial neural network analyses to design an Al–Si casting alloy of minimum porosity, Mater. Des. 27 (7) (2006) 605–609.

[13] A. Shafyei, S.H. Mousavi Anijdan, A. Bahrami, Prediction of porosity percent in Al–Si casting alloys using ANN, Mater. Sci. Eng. A 431 (1–2) (2006) 206–210.

[14] F.F. Xia, C. Liu, F. Wang, M.H. Wu, J.D. Wang, H.L. Fu, J.X. Wang, Preparation and characterization of nano Ni–TiN coatings deposited by ultrasonic electrodeposition, J. Alloys Compd. 490 (2010) 431–435.

[15] M.H. Wu, Z. Li, F.F. Xia, L.Y. Chen, Preparating technique of nano-cermet composite layer with method of ultrasonic-electrodepositing, Mater. Mech. Eng. 29 (8) (2005) 58–61 (in Chinese).

[16] I. Corni, R.J. Chater, A.R. Boccaccini, M.P. Ryan, Electro co-deposition of Ni–Al_2O_3 composite coatings, J. Mater. Sci. 47 (2012) 5361–5373.

[17] X.T. Yuan, Y. Wang, D.B. Sun, H.Y. Yu, Influence of pulse parameters on the microstructure and microhardness of nickel electrodeposits, Surf. Coat. Technol. 202 (9) (2008) 1895–1903.

[18] M.R. Vaezi, S.K. Sadrnezhaad, L. Nikzad, Electrodeposition of Ni–SiC nano-composite coatings and evaluation of wear and corrosion resistance and electroplating characteristics, Colloids Surf. A 315 (2008) 176–182.

[19] L. Benea, F. Wenger, P. Ponthiaux, J.P. Celis, Tribocorrosion behaviour of Ni–SiC nano-structured composite coatings obtained by electrodeposition, Wear 266 (2009) 398–405.