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Application of artificial neural network and adaptive neuro-fuzzy inference system to investigate corrosion rate of zirconium-based nano-ceramic layer on galvanized steel in 3.5% NaCl solution

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ABSTRACT

A nano-ceramic Zr-based conversion solution was prepared and optimization of Zr concentration, pH, temperature and immersion time for the treatment of hot-dip galvanized steel (HDG) was performed. SEM microscopy was utilized to investigate the microstructure and film formation of the layer and the anticorrosion performance of conversion coating was studied using polarization test. Artificial intelligence systems (ANN and ANFIS) were applied on the data obtained from polarization test and the models for predicting corrosion current density values were attained. The outcome of these models showed proper predictability of the methods. The influence of input parameters was discussed and the optimized conditions for Zr-based conversion layer formation on the galvanized steel were obtained as follows: pH 3.8–4.5, Zr concentration of about 100 ppm, ambient temperature and immersion time of about 90 s.

1. Introduction

Galvanized steel is extensively used in industries because of its appropriate properties including high strength and high corrosion resistance. These abilities make galvanized steel a suitable choice for application in building, automotive body parts and water distribution systems [1–3]. Zinc layer protects the steel from corrosion due to galvanic effect and extends the lifetime of steel. Poor adhesion of the organic coating on the surface is one of the most important limitations of the galvanized steel applications. Conversion coatings are applied on the metallic substrates in order to protect them against corrosion and also improve adhesion of the organic coatings. Phosphate and chromate are the most common types of conversion coating which have been used for several years on galvanized steel. Recently attempts have been carried out in order to replace them with other substances, due to their environmental issues and energy saving problems [4,5]. Hexavalent chromium in Cr-based pretreatment solutions is known as a toxic material and its use has been restricted in recent years [5]. Phosphating is a long-lasting process operating at elevated temperatures, resulting in more energy consuming [6]. Moreover sludge removal of the phosphating bath is an expensive process [7]. Extensive researches have been made in order to use novel alternatives instead of conventional conversion coatings. Several investigations on conversion coatings based on different materials including chromium (III) [8,9], vanadium [10,11], cerium [12,13], organo-silanes [14], molybdenum [15,16] and etc. have been conducted. The most promising materials are zirconium and titanium based conversion coatings [17,18]. Zirconium based treatments have been developed for magnesium [19], steel [20–22], aluminum alloys [23] and galvanized steel surfaces [24,25], but most of them have been used as commercial solutions and their exact composition are not available. Optimization of different factors affecting the conversion coating performance has become a serious concern of researches [25]. It has been reported that, electrochemical techniques as well as surface analytical methods have been utilized to optimize the operating conditions [20,25].

Artificial intelligence techniques like artificial neural networks (ANN) and adaptive neuro-fuzzy inference systems (ANFIS) are efficient techniques to predict optimal conditions, because they do not need incorporation of any assumptions or simplifications. These methods attempt to mimic the ability of the human brain to learn patterns. In both methods, the models aim to acquire the relationship between a historical set of model inputs and corresponding outputs. There are some advantages which make them more appropriate than conventional statistical methods in the case of large sets of inputs [26]. These methods do not need

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predefined mathematical equations of the relationship between the model inputs and corresponding outputs and rather use the data alone to determine the structure of the model [27]. This enables of these methodologies to overcome the limitations of existing modeling methods. These methods also offer potentially great flexibility with respect to the ability to approximate a wide variety of functions. Jang [28] presented the ANFIS system in 1993 which was a combination of both neural network and fuzzy logic. It designs the rules based on the input data, and calculate the weights based on neural network design. ANFIS is capable of handling complex and non-linear problems and it operates as a feed forward back-propagation network. Even if the targets are descriptive, ANFIS may reach the optimum result rapidly. There are two learning methods in the neural section of the system: hybrid learning method and back propagation learning method. Fig. 1, depicts the neural network and ANFIS structure with four inputs and one output that have been used in the present work. The other parameters of the model structure such as the number of neurons and number of hidden layers could be determined considering optimization of some variables such as MSE, RMSE or R^2.

Kumar and Buchheit [29] reported prediction of long-time performance of coated samples using ANN. They correlated the failure time of a coating in salt spray test by the parameters obtained from electrochemical impedance spectroscopy with the aid of neural network. Rosen and Silverman [30] utilized an ANN system to predict the type of corrosion occurred on the metal surface through polarization test data. Other researchers, have predicted some parameters such as corrosion rate and pit growth by intelligent systems [31–33]. Bucolo et al. [34] developed a model for corrosion prediction using neuro-fuzzy expert system. Using these intelligent methods for evaluating conversion coating has not been done and there is no systematic work in the literature.

In the present work, the artificial neural network (ANN) and adaptive neuro-fuzzy inference systems (ANFIS) were utilized to model the corrosion rate as a function of pH, zirconium concentration, temperature, and immersion time for a hot dip galvanized steel. Furthermore, the effect of each factor was discussed and optimized treatment conditions were obtained for the conversion coating solution using the mentioned methods.

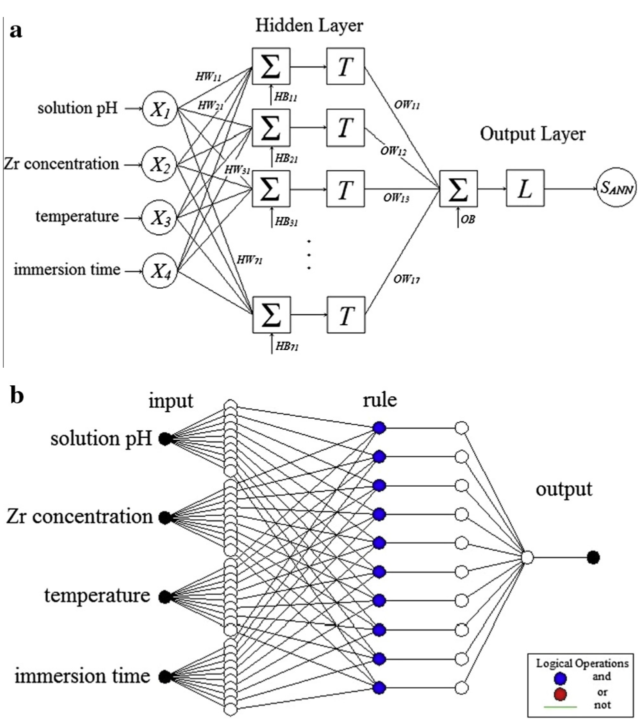

Fig. 1. (a) Structure of ANN model and (b) structure of ANFIS model.

2. Materials and methods

2.1. Surface preparation and zirconium conversion coatings (ZCCs)

Hot-dip galvanized steel sheets with thickness of 1500 µm and zinc layer thickness of about 20 µm were prepared from Foolad Mobarakeh Co. In order to surface preparation prior to conversion coating, the specimens were degreased with commercial acetone, followed by rinsing with deionized water and drying.

Hexafluorozirconic acid solution was purchased from Sigma Aldrich Co. and prepared in different pH values and Zr concentrations. Ammonium bicarbonate (NH_4HCO_3) (0.5% w) was used to adjust pH. The ranges of the studied parameters are summarized in Table 1. Zr treated specimens were then rinsed with deionized water and dried in hot air stream. For variation of each factor, the other three factors were constant (Table 2).

2.2. FE-SEM analysis

The morphology and film formation of conversion layer was investigated using a high resolution FE-SEM MIRA-TESCAN microscope equipped with an Energy Dispersive X-ray Spectroscopy (EDS) system.

2.3. Polarization test measurements

Polarization test was conducted in order to evaluate the corrosion current densities of the galvanized steel samples with and without Zr based conversion coating. The measurements were done by an AUTOLAB PGSTAT10 with a scan rate of 5 mV/s and in the potential range of ±100 mV versus open circuit potential. All samples were sealed with a beeswax and colophony resin mixture, leaving 1 cm^2 area unsealed to expose to 3.5% w/w NaCl solution. The specimens were allowed 60 s in 3.5% w/w NaCl solution to attain a stable open-circuit potential (OCP) before starting the polarization scan. Corrosion current density (i_{corr}) was extracted from polarization curves using Tafel extrapolation technique. The electrochemical system used was including platinum electrode (auxiliary electrode), saturated Ag/AgCl electrode (reference electrode) and metal sheet (working electrode).

2.4. Procedure of modeling data with ANN

A multi-layer perceptron ANN with one hidden layer was used for modeling procedure. For all data sets a "hyperbolic tangent sigmoid transfer function" in the hidden layer and a "linear transfer function" in the output node was employed. 80% and 20% of total experimental data (55 data) were used as training and testing data, respectively (all data were normalized). The ANN was trained using the scaled conjugate gradient back propagation algorithm. One hidden layer was considered and in order to determine the optimum number of neurons in the hidden layer, a series of topologies was used, in which the number of neurons was varied from 2 to 30. Each topology was repeated 10 times to avoid random correlation due to random initialization of the weights and bias. The optimal architecture of the ANN model and its parameters variation were determined based on the minimum value of the root mean square error (RMSE) of the training and testing sets. RMSE measures the performance of the network, according to equation (1):

$$\mathrm{RMSE} = \sqrt{\frac{1}{N}\sum_{i=1}^{i=N}\left[(S_{\mathrm{ANN}})_i - (S_{\exp})_i\right]^2} \qquad (1)$$

where N is the number of data points, $(S_{\mathrm{ANN}})_i$ is the ANN prediction and $(S_{\exp})_i$ is the experimental response. Fig. 2 illustrates the relation between RMSE (network error) and number of neurons in the hidden layer. As it can be seen, the RMSE is minimum in case of 7 neurons. Hence, a two-layered perceptron neural network (with 6 artificial neurons in the hidden layer) has been used for modeling of the corrosion rate.

2.5. Procedure of modeling with ANFIS

Adaptive neuro-fuzzy inference system (ANFIS) has been used to predict the corrosion rate of Zr treated samples. Then, the prediction performance of this model was estimated and compared to ANN. ANFIS can be considered as a network

Table 1
Studied parameters and their ranges.

Parameters	Range
pH	3–5.5
Zr concentration	50–500 ppm
Temperature	25–50 °C
Immersion time	30–120 s