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Prediction of corrosion behaviour of Alloy 22 using neural network as a data mining tool

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ABSTRACT

Objective of this work was to develop an algorithm to predict behaviour of corrosion resistant metal alloys using a supervised neural network method as a data mining tool. We studied corrosion data on a nickel-based alloy, Alloy 22 which is of great industrial interest. This is an extension of a previously reported study on metallic glasses, carbon steel, and grade-2 titanium. The data mining results allow us to categorize and prioritize certain parameters (i.e. pH, temperature, time of exposure, electrolyte composition, metal composition, etc.) and help us understand the synergetic effects of the parameters and variables on corrosion behaviour.

1. Introduction

'Data mining' is the extraction of knowledge and information from large amount of data [1,2]. It has applications in a wide range of subjects, including science and technology. The goal of data mining varies according to the application. We previously reported prediction of corrosion behaviour of metallic glasses, carbon and alloy steel, and grade-2 titanium using artificial neural network (ANN) as a data mining tool [3]. In this paper, we studied Alloy 22, a group of nickel-based alloys that have significant interest in long-term industrial and other applications due to their corrosion resistant behaviour. We presented a general discussion on Data Mining and provided introduction on backpropagation methodology in ANN in the previous paper [3]. We discuss ANN briefly in the present paper, skipping detailed discussion for brevity and focus on presenting the results on Alloy 22 using ANN methodology.

An artificial neural network (ANN) is a computational model based on biological neural networks and consists of an interconnected group of artificial neurons. It can be treated as non-linear statistical data modelling tools that can be used to model complex relationships between inputs and outputs or to find patterns in data. Advantages of an ANN include: (i) an ANN can perform tasks that a linear program cannot, (ii) when an element of the neural network fails, it can continue without any problem by their parallel nature, (iii) a neural network learns and does not need to be reprogrammed, and (iv) it can be implemented in any application without any problem. However, the disadvantages of an ANN include: (i) an ANN needs training to operate and (ii) requires high processing time for large neural networks. Corrosion data on metal alloys are commonly used to obtain unknown parameters in deterministic theoretical models; in order to accurately portray the laws that govern the alloy's behaviour under any environmental condition (generally). Then those models, once those parameters are obtained from data fitting, are used to make "long term" corrosion predictions and prediction on a wider range of environments for the alloy under study. This scientific exercise to create a model and fit measured data to obtain unknown parameters is necessary to develop a predictive methodology that spare us from carrying out infinite measurements – if the metal alloy under study is very good corrosion resistant. Data mining using ANN "short-cuts" the deterministic model and provides a "hidden" map between measured alloy characteristics, environments, and the experimental failure observations on the alloy under research.

Alloy 22 (UNS N06022) is a nickel-based alloy containing by weight: 22% chromium Cr, 13% molybdenum Mo, 3% tungsten W, approximately 3% iron Fe, and the balance Ni [4]. Due to its material composition, Alloy 22 has excellent resistant properties for general corrosion under both oxidizing and reducing conditions [5–8]. General corrosion resistance of Alloy 22 is mainly due to the formation of a thin chromium oxide Cr_2O_3 film and has been studied widely, for example, in Refs. [8–17]. Alloy 22 is also resistant to localized corrosion such as pitting corrosion and crevice corrosion. Critical temperature for pitting and crevice corrosion is typically determined through immersion tests in aggressive solutions. It is possible to initiate localized attack in Alloy 22, but the

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environmental circumstances for doing this are extreme. Of particular concerns are environments of high chloride concentrations at high temperature. Localized corrosion resistance of Alloy 22 has been studied by several researchers; Refs. [6,7,18–29] are to mention a few. Annealed Alloy 22 is highly resistant to environmentally assisted cracking (EAC) in most environments, including acidic concentrated chloride solutions. EAC, stress, fabrication, and other processing effects on Alloy 22 corrosion have been studied as described in Refs. [20,30–36]. Environmental effect on the corrosion properties of Alloy 22 has been the subject of study of several researchers, as presented in Refs. [8,9,37,38].

Sometimes environmental effects on alloy behaviour are not well understood and most of the parameters that play a role on the alloy's potential response are difficult to directly or indirectly measure or quantify. This is particularly correct if the alloys such as Alloy 22 are good corrosion resistant in a given environment and it takes considerable time for them to reach steady state. Because the experimental work domain includes parameters hard to quantify and evaluate, producing data and fitting polynomials to that data in order to extrapolate alloy behaviours over extended period may not be suitable. Data mining, applying to corrosion, represents an assortment of tools available to quantify the response that a given alloy sample will have under a given imposed environment and the effect that the environmental changes have on the alloy sample. Corrosion data on metal alloys are commonly used to obtain unknown parameters in developing deterministic theoretical models, in order to accurately portray the laws that govern the alloys' behaviour under any environmental condition [10,13,14,39–42]. Then those models, once those parameters are obtained from data fitting, are used to make "long term" corrosion predictions and prediction on a wider range of environments for the alloy under study. This scientific exercise to create a model and fit measured data to obtain unknown parameters is necessary to develop a predictive methodology that spare us from carrying out infinite measurements – if the metal alloy under study is very good corrosion resistant, as in the case of Alloy 22. Data mining "short-cuts" the deterministic model and provides a "hidden" map between measured alloy characteristics, environments, and the experimental failure observations on the alloy under research. In the data mining we carried out, special emphasis was given to categorizing the importance of the role of different parameters on the corrosion rates.

Neural network has been applied in the analysis of mechanical properties of metals/alloys [43–50]. However, the application is limited in corrosion studies and analysis, especially for Alloy 22. In our analysis, we studied corrosion rate and weight loss data [51], crevice corrosion data [52] and AC impedance data [41,42] on Alloy 22 for data mining using ANN backpropagation methodology to train NN models. These ANN models were used for comparison and future prediction of the alloy's corrosion behaviour.

In this paper, the data collected is described in Section 2. Database mining of general corrosion data using neural network is discussed in Section 3. Database analysis and results obtained from data mining of localized corrosion data are presented in Section 4. Summary on the data analysis is presented in Section 5 of this paper.

2. Database mining: data collection, database analysis, and knowledge extraction

We have collected corrosion related data on Alloy 22 with different corrosion behaviour. Data presented in this paper were collected from mainly two sources: (i) Publicly available (publications, etc.) and (ii) Personal communication and were divided into (a) general corrosion data and (b) localized corrosion data. For the analysis, we have selected the following sources:

(a) General corrosion:
   i. Alloy 22 [51] data. A sample is shown in Table 1.
(b) Localized corrosion:
   i. Crevice repassivation potential data on Alloy 22 [52]. a sample is shown in Table 2.
   ii. Impedance corrosion data (AC) at different applied potentials and operating conditions on Alloy 22 [41,42]. Sample data are shown as plots in Fig. 3.

We summarize in the following sections a sample of the data we have collected and used for our analysis in this work. The data presented in the tables/graphs are only an example of data collected and not an exhaustive example of all the collected data. Data from multiple experiments, figures and tables that represent the same corrosion variables were integrated into a single database for further analysis.

After pre-processing the data, we trained ANN models for each set of the data using backpropagation methodology and to predict crevice corrosion potential, corrosion rates and temperature effects and to categorize corrosion parameters. Detailed analysis of the general and localized corrosion data are presented in the next sections. In general, the ANN backpropagation algorithm we applied here normalizes the data by default. Bootstrapping and/or several folds of validation are commonly applied to minimize overtraining of data and models. We here applied the validation technique for this purpose.

3. Database mining of general corrosion data on Alloy 22

3.1. General corrosion study using weight loss data [51]

These data on Alloy 22 were collected from Hua et al. [51] and included the environment, type of metal alloy (annealed or welded), temperature, and metal dissolution rate under the specified condition and at a given time. Table 1 shows the weight loss measurements of Alloy 22 in Basic Saturated Water (BSW)-12 at different temperature values. BSW-12 is similar to the environment in the Yucca Mountain area which was of great interest for high level nuclear waste (HLNW). Repository development and Alloy 22 was a highly recommended canister material for nuclear waste disposal due to its high corrosion resistance [8,9,17,20,22,25,29,33,51–61].

Weight loss measurement is the most direct way to measure corrosion loss of metals/alloys. However, the problem with the weight loss measurement is that it takes long time to measure the corrosion loss for that period of time. Therefore, if we can establish the functions that map the corrosion variables to the corrosion loss using the direct measure and predict the corrosion behaviour to the future, i.e. evolution of corrosion, it can be used in the selection of metals/alloys at the environments and conditions of interest. However, some other indirect methods of measurement such as polarization behaviour, impedance study, pitting evolution can be used to complement the direct measures and help predict corrosion behaviour in the far future, validate deterministic models, and find the most appropriate metals/alloys for the applications of interest.

Using the Alloy 22 weight loss data collected from Hua et al. [51], the experimental environment conditions, temperature values in the present case, were mapped to the corrosion rate (weight loss). NN backpropagation method, a supervised learning technique in ANN [3,62–64], was used to fit a model to the collected experimental data using NeuralWare Software [65]. The topology of the ANN was as follows: 4 inputs (temperature values in degrees Celsius, sample treatment types: welded or annealed, and the exposure time, which was a constant in the current example),

Table 1
Measured values of corrosion weight loss of annealed and welded Alloy 22 in basic saturated water (BSW)-12 at different temperature values. Data source: Hua et al. [51].

Environ	Temp (°C)	Material condition	Exposure time (h)	Weight loss (g)	Surf area (cm^2)	Corrosion type
BSW-12	60	Annealed Alloy 22	1344	0.00065	22.79074	General
BSW-12	70	Annealed Alloy 22	1344	0.00136	22.79074	General
BSW-12	100	Annealed Alloy 22	1344	0.00221	22.79074	General
BSW-12	105	Annealed Alloy 22	1344	0.00222	22.79074	General
BSW-12	60	Welded Alloy 22	1344	0.00076	22.79074	General
BSW-12	90	Welded Alloy 22	1344	0.00176	22.79074	General
BSW-12	100	Welded Alloy 22	1344	0.00178	22.79074	General
BSW-12	105	Welded Alloy 22	1344	0.00265	22.79074	General

two hidden layers consisting of 5 neurons each, and 1 output (corrosion weight loss in mg). A hyperbolic tangent (tanh) function was used as the transfer function in the ANN model. The Levenberg–Marquardt algorithm was used as the optimization algorithm and the error minimization tolerance was set to 1%. Once the NN backpropagation output was "thought" to "mimic" the experimental results, the NN was said to have learned the mapping function between the inputs and the output. The process of learning was obtained by adapting the weights or connections between the neurons contained in the different layers used to design the neural net. Then, the NN backpropagation was ready to be tested. In the testing mode, the weights or neuron connections were not changed and for each input vector, the net reproduced an output. It should be noted that the results can be reproduced with different ANN models with different hidden layers and/or different number of neurons, given the input–output, and error stopping criteria. In other words, the ANN may not be unique, which is the nature of ANNs. In our analysis, we ran the NN model with different hidden layers and with different sets of neurons in each layer. The selected model with 2 layers having 5 neurons each was the smallest network of all that were tested and provided the desired error tolerance. Prediction of weight loss using the temperature values as the input was validated using test data.

The results of the analysis are shown in Fig. 1. The corrosion weight loss of annealed and welded samples of Alloy 22 was plotted vs. temperature in Fig. 1a. The NN model satisfactorily 'mimicked' the corrosion weight loss of Alloy 22 in the range of temperature the NN was trained. Not much difference in the weight was observed for the difference in material conditioning (annealed and welded). It was noted in the data source [51] that, during the investigation of samples, no difference was observed in terms of crevice or pit growth on the samples. Although some noisy effect in the weight loss was observed in the experimental measurement, it was not observed in the NN results. R^2 (Square of the Pearson Product–Moment Correlation Coefficient) value between experimental data and the NN results was used as a measure of goodness of fit and was calculated to be 80%. Cross-validation was used to reduce data over-fitting. It should be noted that due to the limited experimental data values from this particular source, we used the same data for training and testing in cross-validation. However, if available, more data points would be valuable to test the efficacy of the model with data not seen by the model during training. In that case, several folds of cross-validation can be applied by dividing the data into randomly selected segments and the validation error can be averaged over the validation folds, which would provide a more robust model.

The NN results show consistent temperature effect on the weight loss of the samples. We investigated whether corrosion rate (CR) followed Arrhenius rule ($k = Ae^{-E_a/RT}$), where k is reaction rate, A is the pre-constant or intercept, E_a is activation energy, R is gas constant, and T is temperature in absolute values. CR was calculated from the weight loss, duration of experiments, surface area of the samples, and the density of the samples. When we plotted Ln(CR) vs. $1/T$, we indeed observed a straight line with negative slope of 3.38 for the NN results, as shown in Fig. 1b. The validity of the Arrhenius rule makes the model applicable in the general sense of thermal properties of reaction kinetics of corrosion of metals/alloys. Ashida et al. [66] investigated the temperature effect on passive corrosion of Alloy 22 by using a temperature-oscillating heated electrode technique (TOHET) and showed similar relations in terms of corrosion potential and current density. With the model developed in this work i.e. the Arrhenius parameters, corrosion rates at temperature values outside the range that the model was trained can be predicted. However, more data points would strengthen such a conclusion.

4. Database mining of localized corrosion data on Alloy 22

4.1. Crevice corrosion study using crevice repassivation potential on Alloy 22 [52]

We have collected data on Alloy 22 as shown in Table 2. The experimental data collected by Dunn et al. [52] included material condition (welded and annealed), environment (temperature, pH, chloride concentration), sample treatment (welded, annealed,

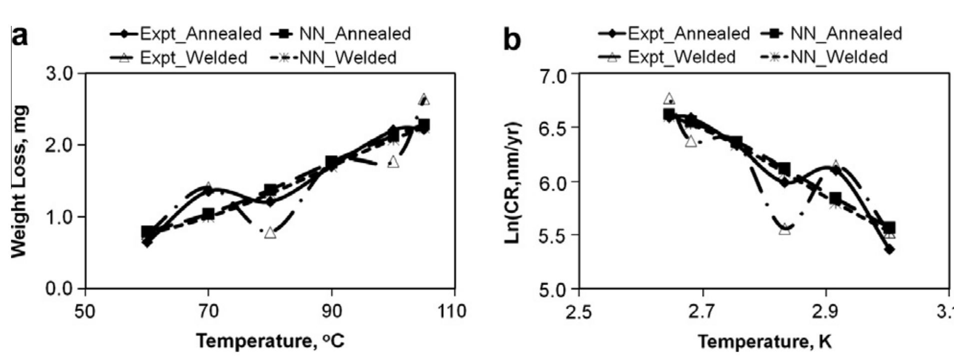

Fig. 1. Comparison of experimental values and neural network results for annealed and welded Alloy 22 in BSW-12 at different temperature values with exposure time of 1344 h and surface area of samples of 22.79 cm^2. (a) Corrosion weight loss vs. temperature, T; (b) corrosion rate (CR) vs. $1/T$. Data source: Hua et al. [51].

aging time), type of corrosion, and crevice repassivation potential. The alloy would be expected to resist localized corrosion if the corrosion potential is cathodic (active) with respect to the repassivation potential. One way to choose the repassivation potential is the potential at which the anodic forward and reverse scans cross each other. Alternatively, it can be chosen as the potential at which the current density reaches its lowest readable value on the reverse portion of the polarization scan. The reason for the second approach is that for some polarization scans, the forward and reverse portions of the polarization scan may not cross each other. In other words, crevice repassivation potential plays critical role in determining the resistance of the alloy under consideration in the environment of interest.

We here developed a NN model that maps the environment, material condition, and sample treatment to the crevice repassivation potential by means of a supervised neural network mapping. The model can be used as a predictive tool to investigate the material susceptibility to localized corrosion due to alloy treatment and different environmental conditions (pH, and [Cl^-]).

Fig. 2 shows the performance of the NN mapping such that the model understood the underlying functions of the mapping of the environmental condition and metal treatment to the corrosion resistance of Alloy 22. The topology of the ANN was as follows: 8 inputs (sample treatments: welded or annealed, temperature, pH, [Cl^-], aging time, types of corrosion i.e. general or localized), two hidden layers consisting of 5 neurons each, and 1 output (repassivation potential). The selected ANN model with 2 layers each having 5 neurons was the smallest network of all that were tested, providing the desired error tolerance. The highest contribution to the NN predictions of crevice corrosion potential from the input variable with the highest weighting was the chloride concentration (input # 5 in Fig. 2b: input contribution) which is very realistic. The second most contributing input was the temperature (input # 3 in Fig. 2b: input contribution) at which the experiments were conducted. R^2 value between experimental data and the NN results was calculated to be 100%. Two fold cross-validation was used to reduce data over-fitting. In each fold, the NN model test set included 20% data points not seen by the NN model while trained with the rest of the 80% data. The validation error was averaged over the folds.

Since Alloy 22 is one of the most corrosion resistant Ni-alloys, it is important to study how localized corrosion affects Alloy 22 under severe conditions, for example, in high chloride concentrations. The crevice repassivation potential is one of the important indicators of pitting/crevice corrosion behaviour including metastable pitting, etc. Therefore, it will be valuable to gather more data on Alloy 22 to further study crevice corrosion/pitting behaviour.

4.2. *Localized corrosion, AC impedance data on Alloy 22 [41,42]*

For general corrosion or when the localized corrosion process is slow, the technique of choice is AC impedance spectroscopy. AC impedance data are often used to develop deterministic models or equivalent circuits to predict corrosion behaviour of metals/alloys i.e. when direct corrosion measurements such as weight loss, etc., are not available or expensive or time consuming for the time period or environments of interest. The corrosion behaviour can be determined from the predictive, deterministic models. In a different method, AC impedance data (Nyquist plots) are used to calculate the polarization resistance and in turn, the corrosion rate by determining the real impedance value at which the imaginary impedance intersects the real axis at low frequency.

AC impedance data received from the SRI International [41,42] were analyzed to study the localized corrosion (pitting) behaviour under high applied potential. Fig. 3 is an example of impedance or Nyquist signatures measured in order to learn the localized corrosion process that takes place in Alloy 22 in a saturated NaCl electrolyte at 80 °C. The experimental conditions were: saturated NaCl in pH 3 HCl, 80 °C, de-aerated conditions, Alloy 22 (as received) with surface area 6.45 cm^2 at applied voltages: 550 mV, 700 mV, 800 mV, and 900 mV vs. SCE. We observed a large difference between Fig. 3a and b, where only the polarization potential was changed. The loops in the Nyquist plot at low frequency is interesting and important, because this behaviour is not observed at applied potential less than 500 mV_{SCE} and the rest of the conditions remaining the same. As per the experimental evidence [41,42], pitting was observed at high applied potential.

It is the low frequency impedance measurement that contains very useful information from corrosion viewpoint. For example, the polarization resistance that is used to determine the general corrosion rate is obtained by determining the real impedance at which the imaginary impedance value intersects the real axis at low frequency. It is sometimes impossible to measure the low frequency tail of the impedance (for example, a datum point at 10^{-4} Hz will take 10^4 s to measure). Accordingly, it is of great interest to be able to learn the impedance frequency dependence of a system and use a smart extrapolation to extract the information available. For example, if a NN model is developed that learns the mapping functions of impedance–frequency dependence of a system, it can be used to predict the polarization resistance and hence the corrosion rate of the system at the conditions of interest.

An NN model was developed using the frequency dependent impedance data and was validated using experimental data. The topology of the ANN was as follows: 2 inputs (frequency (w) and log(w)), two hidden layers consisting of 100 neurons each, a third hidden layer with 10 neurons, and 4 outputs (Zr and Zi i.e. real and imaginary impedances, respectively, abs(Z), and Phase). The topology of the ANN model in this case was selected randomly that provided the desired level of error tolerance. The model is in good agreement with the experiment, as shown in Fig. 4. It should be noted that due to the limited experimental data, we used the same data for training and testing in cross-validation. However, if more data points are available, several folds of cross-validation can be applied by dividing the data into randomly selected segments and the validation error can be averaged over the validation folds, which would provide a more robust model. The developed model

Table 2
Experimental values of crevice repassivation potential on Alloy 22. Data source: Dunn et al. [52]. ASW—'as welded', MA—"mill annealed".

Material condition	Temp (°C)	pH	[Cl^-] (M)	Aging lime at 870 °C (min)	Ercrev (mV vs. SCE)	Corrosion type
ASW	95	8	0.5	0	279.56	Localized
ASW	95	8	1.0	0	302.45	Localized
MA	95	8	1.0	0	317.47	No local corrosion
MA	95	8	4.0	0	13.67	No local corrosion
ASW	125	8	1.0	0	−92.05	Localized
ASW	125	8	4.0	0	−226.55	Localized
MA	95	8	4	0.5	−248	Intergranular (local)
MA	95	8	4	4	−255	Intergranular (local)
MA	95	8	4	24	−257	Intergranular (local)

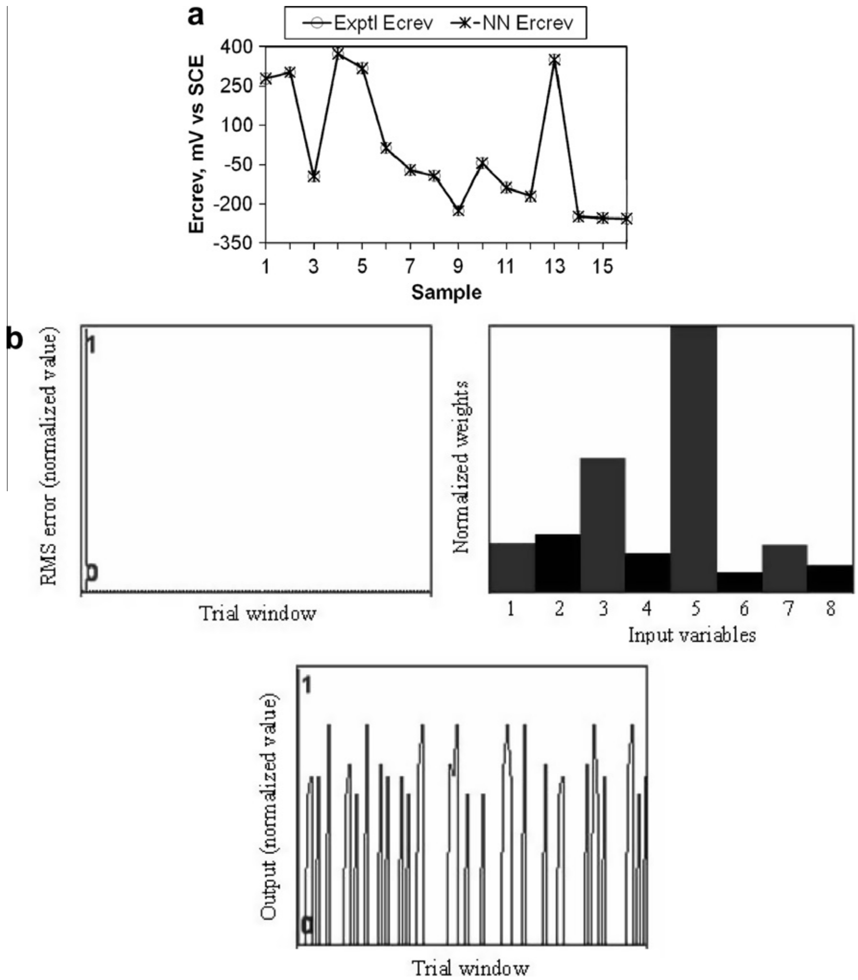

Fig. 2. (a) Comparison of experimental values and neural network results for crevice repassivation potential of Alloy 22. Experimental condition: pH = 8, varying [Cl$^-$] concentrations (0.5–4 M) and temperature (95–125 °C) as shown in Table 2. (b) A sample of the NN model that "mimicked" the experimental data.

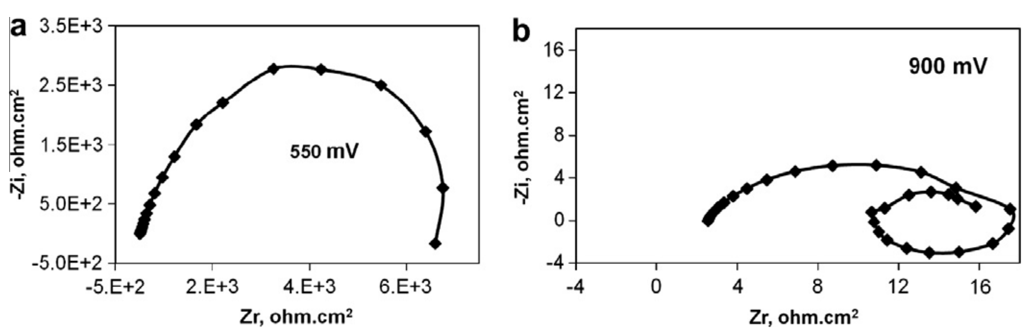

Fig. 3. Nyquist Plot for Alloy 22 experimental data with a sample surface area of 6.45 cm^2 at (a) 550 mV$_{SCE}$ and (b) 900 mV$_{SCE}$. Experimental condition: Saturated NaCl in pH = 3 HCl, 80 °C, de-aerated solution [41,42].

was used for impedance prediction at user generated frequencies and the results are shown in Fig. 4(a–d). We also developed NN models for different applied potentials (550 mV, 700 mV, 800 mV, and 900 mV vs. SCE) using a different set of inputs i.e. different functional forms of the frequencies (sqrt(w), w^2, w^3, and log(w)) and impedances (Zr and Zi, abs(Z), log(abs(Z)), and Phase). The results came out in good agreement with experimental data, similarly as presented in Fig. 4. The goodness of fit in terms of the Pearson's correlation (R^2) values between the experimental data and NN results were 99% and above for the impedance values shown in Fig. 4.

We are in the process of developing models for the temperature and the applied potential dependence and will report the results in the near future. Future research plan also includes development of deterministic models for the prediction of Alloy 22 corrosion behaviour, the temperature dependence, and passivation-repassivation properties and compare the results with NN results.

5. Summary

A supervised neural network (NN) mapping using 'backpropagation' method was developed for Alloy 22 such that the NN model

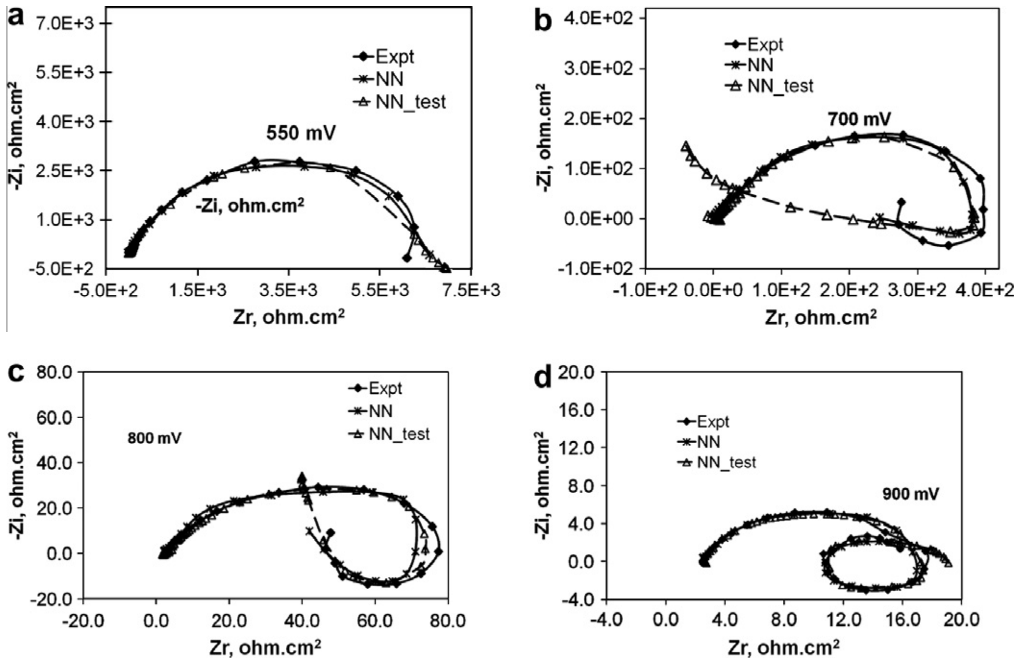

Fig. 4. Nyquist plot for Alloy 22 at pH = 3, T = 80 °C in Saturated NaCl for a sample with surface area of 6.45 cm^2 at applied potentials of (a) 550 mV$_{SCE}$, (b) 700 mV$_{SCE}$, (c) 800 mV$_{SCE}$, and (d) 900 mV$_{SCE}$. Expt: experimental data, NN: neural network model prediction using the same input data that was used for training, NN_test: neural network model predicted impedance at user generated test frequencies.

learned the underlying functions in mapping corrosion variables and parameters to different corrosion metrics (corrosion rate, crevice repassivation potential, impedance values, etc.).

Corrosion data on Alloy 22, a highly corrosion resistant alloy were collected from mainly publicly available articles and personal communication. Both general corrosion data and localized corrosion data were collected, pre-processed (transformed, cleansed, integrated, and normalized) and used in the NN analysis.

Direct measurements of corrosion weight loss data for Alloy 22 were used in the NN mapping to predict future corrosion weight loss under similar environmental and sample conditions and validation of the NN model using experimental data available showed good agreement. Temperature effect on corrosion rate was shown to follow Arrhenius rule. Crevice and pitting corrosion behaviours in terms of repassivation potential of Alloy 22 were predicted using the NN model that showed excellent agreement when validated with experimental data. AC Impedance data revealed interesting corrosion behaviour of Alloy 22 that, under applied potentials >550 mV$_{SCE}$, significant pit growth was observed during experiments [41,42] on the highly corrosion resistant Alloy 22 samples. Developed NN models using the AC impedance data predicted this behaviour quite efficiently when validated using experimental test data sets and user-generated frequency data sets. Future research plan includes development of deterministic models for the prediction of Alloy 22 corrosion behaviour, the temperature dependence, and passivation–repassivation properties and compare the results with NN results.

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