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Computer Simulation Using Fuzzy Logic Model to Predict Hot Corrosion Kinetics in Molten Salt of Steel-T21 Coated by Simultaneous Yttrium-Doped Aluminizing-Siliconizing process

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Abstract

The present paper describes fuzzy logic simulation of an experimental study on the behavior of hot corrosion in molten salt (Na_2SO_4) of steel-T21 coated by simultaneous yttrium-doped aluminizing-siliconizingprocess . Diffusion coating was carried out at $1050^{o}C$ for 6 hr under Aratmosphere . The weight change measurements made on the coated steel during the cyclic tests are used to determine kinetics of hot corrosion at temperature range ($800-1000^{o}C$) for 100 hr at 5 hrcycle .X-ray diffraction and optical microscope are used to characterize the oxide phases where the oxide phases that formed on coated system are SiO_2 and Al_2O_3 .The parabolic rate constants (K_p) calculated show that the corrosion rate is minimum at $800^{o}C$ compared to other temperatures. The experimental results, the fuzzy logic model, and the statistical results showed good correlations.The fuzzy logicmodels are developed using Matlab toolbox functions.

Keywords: Hot Corrosion, Steelt21, Fuzzy-Logic Controller

المحاكاة الحاسوبية بإستخدام نموذج المنطق المشّوش للتنبأ بحركيات التآكل الساخن في الملح المنصهر للفولاذT21 المطلي بطريقة الألمنة- سلكنة الآنية المحورة باليتيريوم

الخلاصة

يتضمن هذا البحث إستخدام المحاكاة الحاسوبية بواسطة المنطق المشّوش للدراسة التجريبية حول سلوك التآكل الساخن في الملح المنصهر (Na_2SO_4) للفولاذ نوع-T21 المطلي بإستخدام طريقة الألمنة- سلكنة الآنية المحورة بواسطة اليتيريوم . و تم تنفيذ عملية الطلاء الإنتشاري عند درجة $1050^{o}C$ و 6 hr تحت أجواء الآركون . و تم إستخدام قياسات التغير بالوزن التي أجريت على الفولاذ المطلي خلال الإختبارات الدورية لتحديد حركيات التآكل الساخن عند مدى درجات الحرارة ($800-1000^{o}C$) لمدة 100 hr (5 ساعة لكل دورة) . و أستخدم كل من حيود الأشعة السينية لتحديد الأطوار الأوكسيدية المتكونة على نظام الطلاء ، حيث لوحظ تكون الأطوار Al_2O_3 و SiO_2 . و أظهرت نتائج ثوابت معدل القطع المكافئ K_p ان معدل التآكل يكون في الحد الأدنى عند درجة $800^{o}C$ مقارنةً مع الدرجات

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الأخرى . كما لوحظ التطابق الجيد مابين النتائج التجريبية، نتائج نموذج المنطق المشوش و النتائج الإحصائية . هذا، و قد تم تحديد نموذج المنطق المشوش بواسطة صندوق أدوات برنامج ماتلاب .

INTRODUCTION

The behavior of high temperatures has been significant in the improvement of society for many countries. Structural equipment in many high technology areas has to be operating under severe circumstances of temperatures, pressure and corrosive environment. Consequently, materials degradation at high temperature is a severe trouble in several industries. Coal is a compound and comparatively polluted fuel that contains varying quantity of sulphur and a considerable fraction of non combustible mineral constituents, commonly called ash. The vast scientific literature available is evidence that corrosion and deposits on the firesides of boiler surfaces or in gas turbines represent important problems. Metals and alloys may experience accelerated oxidation when their surfaces are coated by a thin film of fused salt. This mode of attack is called hot corrosion , and the most dominant salt involved is Na_2SO_4. High temperature degradation is one of the main failure modes of hot-sections components in the gas turbines , so understanding corrosion and wearing down by fly ashes and unburned carbon particles are the man trouble to be solved in these applications . Therefore, the increase of high temperature oxidation (Hot Corrosion) protection systems in industrial turbines is a very important topic from both engineering and an economic perspective [1].Low alloy steels are generally considered to comprise plain carbon steels and steels with a total alloying content of up to 12%. As such, they are much cheaper than more highly alloyed alloyed materials and are often used in large quantities in heavy engineering industries. Whilst these materials are not yttrium generally selected for resistance to high temperature corrosion (the material choice is largely dictated by cost, ease of fabrication and mechanical properties) they are often required to operate in high temperature aggressive environments. For instance, the power generation, refuse incineration and chemical process industries use many miles of low alloy steel heat exchanger tubes. Hence, the high temperature oxidation properties of low alloy steels are often important in determining component life [2].The development of a surface oxide scale limits the degradation of a pure metal or alloy in a hot oxidizing environment. The addition of reactive elements which have a high affinity for oxygen (such as Y, Ce, Hf) may further improve the oxidation resistance through various effects [3]:

- Promotion of the selective oxidation of an element which forms a stable oxide of low diffusivity (such as Al_2O_3 and SiO_2).
- Reduction of the growth rate of oxide scale.
- Inhibition of scale failure (i.e. through thickness cracking and scale/substrate interfacial decohesion) [1].

Reactive-Element (RE) additions may be provide either as metallic or oxide dispersoide components in bulk alloy, or as surface produced by coating. They are used predominantly with Cr_2O_3- and Al_2O_3-forming alloys to resist aggressive environment [4]. V.Provenzano and coworkers [5].proposed a model of mechanical keying due to the formation of oxide pegs rich in active elements, the role of these peg being to anchor the oxide scale to the coating alloy. It was found that the addition of yttrium prevents the sulfur segregation to the alloy/scale interface, either by reacting with sulfur to form a stable sulfides or by tying up the sulfur by segregation to internal oxides surfaces [6]. Thus, the addition of small amount of reactive element [Y,Ce, La, Hf, Zr, Th] to an alloy resulted in substational improvements in the adherence of their oxide scales during thermal cycling [7]. Previous work [8,9,10,11,12], has shown that such reactive element additions are effective in improving the high temperature corrosion of iron-base alloys by improving the resistance of protective scales to spallation.It was found that the scale formed on germanium -free alloy is typically convoluted or wrinkled and poorly adherent, the germanium -containing alloy produces a flat and adherent oxide. In recent years germanium has become the most commonly used of these reactive elements. The amount of the reactive element needed to produce the beneficial effect is small, (typically 1wt.% or less). Heat-resisting alloys depend on the formation of a protective oxide on the metal surface to limit section loss by oxidation. Spallation of oxide may involve fracture in the oxide adjacent to the metal surface, fracture in the metal immediately below the interface, or by separation at the interface itself. In the last case, failure involves not only the magnitude of the stresses, but also a consideration of the interfacial adhesion.

This paper proposes a fuzzy logic model for the hot corrosion kinetics of coated system consists of deposit of yttrium-doped aluminum and silicon on the surface of steel-T21 alloy using single step pack cementation to enhance the hot corrosion resistance in steel-T21 . The cyclic oxidation behavior of coated steel-T21 alloy will be studied in the temperature range 800-1000°C .

EXPERIMENTAL PROCEDURE COATING SYSTEM

The substrate alloy used in this study was low alloy steel (Type T21-ASTM A200-94). The nominal composition and the chemical analysis of low alloy steel are shown in Table (1) and Table(2) respectively. The low alloy steel samples were cut into squares shapes with dimensions (20mm× 20×mm×5mm) with small hole of 2mm diameter was drilled in each sample for holding. All surfaces, including the edges were wet ground using 120, 220, 320, 600, 800, and 1200 grit silicon carbide papers. These samples were then cleaned with water, degreased with acetone, and then ultrasonically cleaned for 30 minutes using ethanol as a medium. After drying, the samples were stored in polyethylene zip-lock bags. The dimensions of all samples were measured. The pack mixture used for aluminum-silicon diffusion coating consisting of 16 Wt.%Al powder (50-60 μm in particule size) as an aluminum source, 6 Wt.%Si powder (50-60 μm in particule size) as a silicon source, 2Wt.% NaF and 2Wt.%NaCl as activator and the balance was silica-powder (70-120 μm in particule size). All pack powders was sized by sieving method and 1Wt.% of the pack silica filler was replaced by yttrium . Low alloy steel was placed in a sealed stainless steel cylindrical retort of 50mm in a diameter and of 80mm in a height in contact with the pack mixture. The retort was then put in another stainless steel cylindrical retort of 80mm in a diameter and 140mm in a height. The outer retort has a side tube through which argon gas passes and second in the top cover for argon gas outlet. Type-k calibrated thermocouple was inserted through the cover of the outer retort for recording real temperature near inner retort.. Pack cementation process was carried out at 1050ºC for 6 h under an Ar atmosphere. After coating, the samples were ultrasonically cleaned , and weighed. It was found that the diffusion coating time of 6 h at 1050ºC give a coating thickness of 65-66 μm (using coating thickness gauge type-QuaNix® 1500 , Materials Eng. Department/University of Technology) .

HOT CORROSION TEST

For hot corrosion tests, a 100 wt. % Na_2SO_4 powder was selected as a corrosive salt. Samples were deposited with 100 wt.% Na_2SO_4 concentration until a total coating weight of 5 mg/cm^2 was reached according to A.Anderson and S.Ramachandran, procedure [1] .The samples were measured and weighed first , then placed on a hot plated heated to 110ºC. An air gun sprayed on the saturated aqueous –salt solutions in air mist and a coat of fine salt particles formed on the samples surfaces after the mist settled and the water evaporated. The process was repeated until the dry particles were deposited up to 5 mg/cm^2. Hot corrosion test was performed in a static air at temperarture range (800ºC-1000ºC) for 100 hr at 5 cylces in a programmable tube furnace (Locally-Manufactured/Dep. of Production Eng.& Metallurgy, University of Technology). After testing the samples were cleaned in an ultrasonic bath for 15 min., first in distilled water and then in ethanol. They were then weighed on a digital balance to determine the change in weight.

Microstructure

The microstructure of samples subjected to hot corrosion was examined using optical microscope.

X-Ray Diffraction

A Riga Ku X-Ray generator with Cu Kα radiation at 40 KV and 20 mA was used . The X-Ray is generated by general electric diffractometer , type Philips (pw 1840) , operating at scanning speed of 6º (2 θ) per minute . The detector was moved through an angle of 2θ =10 to 80 degree. The XRD analysis was carried out at S.C. Geological Survey and Mining .

Fuzzy Logic Model

A general structure of fuzzy interference system is shown in Figure (4). Fuzzy logic model for hot corrosion prediction is developed in different stages. The first step in the development of fuzzy logic model is to take the inputs and determine the degree to which they belong to each of the appropriate fuzzy sets via membership's functions as shown in Figures (5). In the fuzzy logic system, the input is always a crisp numerical value limited to the universe of discourse of the input variable. The input crisp variables are time to exposure to hot corrosion salts and temperature Table (5) . The output is a fuzzy degree of membership in the qualifying linguistic set. The fuzzy logic system is based on rules and each of the rules depends on resolving the inputs into a number of different fuzzy linguistic sets. Before the rules are evaluated, the inputs are fuzzified according to each of these linguistic sets. The inputs are fuzzified and the degree to which each part of the antecedent is recommended for each rule . Every rule has a weight (a number between 0 and 1) which is applied to the number given by the antecedent . Once proper weighting has been assigned to each rule , the implication method is implemented . The result is a fuzzy set represented by a membership function , which weights the linguistic characteristics that are attributed to it. The aggregates of a fuzzy set encompasses a range of output values (Weight Gain) and so must be defuzzified in order to resolve a single output value from the set [13] .

RESULTS AND DISCUSSION CYCLIC HOT CORROSION

Table (3) shows data extracted from hot corrosion tests. Figure (1) a shows the weight gain/unit area for molten salt (Na_2SO_4) environment for steel T21 coated with simultaneous Y-doped aluminizing-siliconizing coating system . In molten salt environment , the substrate coating system shows higher weight gain at 1000°C as compared to 800 and 900°C which can be attributed to intense spalling and sputtering as shown in Figure (2), that shows the cross section view images of LOM of the coated system used in this study coated low alloy steel substrate at temperature range between 800-1000°C . From the surface appearance of the samples, the spalled areas are considerably low. It is possible that a small amount of yttrium had segregated on grain boundaries in the scale, and little voids are observed near the alloy/scale interface as shown in Figure (2). The XRD pattern shown in Figure (3) for coated system subjected to molten salt environment at 800,900,and 1000°C after 100 hr cycles indicated the formation of Al_2O_3 and SiO . The formation of this phases in protective scale of the the coatings was suggested to induce requisite resistance for steelT21, i.e. these phases are found to be effective in decreasing corrosion rate in molten salt due to the formation of protective oxide scales Al_2O_3 and SiO [14]. Since yttrium could not be detected with X-ray diffraction at the surface of oxidized samples, the suspected enrichment levels for yttrium segregation must be very low. The addition of yttrium had no visible effect on the external scale morphology developed during the cyclic oxidation of yttrium -doped aluminizing-siliconizing.

Coating system appears good oxidation resistance. First, the most desirable SiO_2, and Al_2O_3 protective scale layer forms on the sample surface during oxidation, and these scales layer has sufficient adherent with the substrate to withstand the imposed cyclic heating and cooling environments. This evident from the continuous weight gain noted during oxidation. Second, the (Al+Si) phase of the coating remaining untransformed totally over the entire exposure period indicates that the Al,Si loss from the coating during oxidation is very slow because of the formation of a spall-resistant silica, alumina layer especially at higher temperatures. The presence of the reactive elements such as yttrium affects the high temperature oxidation in three main ways [12]:

- Increase in the selective oxidation of the elements forming the scale (Al,Si) at the beginning of the oxidation process. A lower content of these elements (16 Wt.%Al powder as an aluminum source, 6 Wt.%Si as an silicon source from the total weight of pack-powder as indicated in experimental procedure) as a silicon source) are needed to produce a continuous protective SiO_2, and Al_2O_3 layers.

- Reduction in the scale growth rate at higher temperature (800-1000°C) by means of altering the transport mechanism in the oxide. The outward diffusion of Al changes to inward diffusion of O^{-2}.
- Increase in the scale to alloy adherence.

The weight gain square (mg^2/cm^4) vs. time or (number of cycles) plot is shown in Figure1b to establish the rate law for the hot corrosion . It is observed from the graph that coated system at (800-900°C) follow nearly parabolic rate law. The parabolic rate constant, Kp, was calculated by linear least-square algorithm to a function in the form of [15]:

$$(W/A)^2 = K_p\, t \qquad \dots (1)$$

Where *W/A* is the weight gain per unit surface area (mg/cm^2) and t indicates the number of cycles representing the time of exposure. The parabolic rate constants for coated system in molten salt were calculated on the basis of 20 cycles data and are reported in Table (4) . The parabolic rate constant for coated system at 1000 °C is found to be greater than the other temperatures.

SIMULATION OF FUZZY LOGIC MODEL

In this study, the fuzzy model has been developed based on 20 experiments of hot corrosion parameters. The fuzzy model was simulated for test cases which has been done within the range of fuzzy set (two inputs and one output) i.e. the simulation is based on the relationship that exits between time, temperature (inputs) and hot corrosion kinetics (output). The purpose of the simulation was to minimize the error of outputs for test case experiments. The measured and predicted values of weight gain are given in Table 6 and Figure 6. The results from fuzzy logic simulation indicated that the predicted values and experimental values closely agreed. The parabolic rate constants, KP calculated from experimentally observed and predicted values are given in Table (7). It can be observed that there is good agreement between experimental and predicted values.

Statistical Analysis

The root mean square error (RMSE) was used to estimate the variation, expressed in the same unit as the data between simulated and measured values. This parameter is defined by [16]:

$$RMSE = \left(\sum_{i=1}^{n} (S_i - M_i)^2/n\right)^{1/2} \qquad \dots (2)$$

Where M_i and S_i are the measured and simulated values respectively, for *i*th data point of *n* observations. The RMSE of fuzzy modeling analysis is presented in Table (8) for the test cases. As shown form this table that all values of RMSE are less than one i.e. there is good agreement between the experimental and predicted values .

CONCLUSIONS

The conclusions resulting from the current study are summarized as follows:

- Steel T21 coated with simultaneous Y-doped aluminizing-siliconizing coating system is resistant to hot corrosion at temperature range (800-1000^{o}C) .
- The parabolic rate constants (K_p) for steel T21 coated with simultaneous Y-doped aluminizing-siliconizing coating system at temperatures 800, 900 and 1000^{o}C are 2.4×10^{-4}, 6×10^{-3} and $2.4\times10^{--2}$ respectively.
- Steel T21 coated with simultaneous Y-doped aluminizing-siliconizing coating system has higher corrosion resistance at 800^{o}C as compared to other two temperatures.
- The formation of protective oxide scales such as Al_20_3 and SiO_2 is responsible for imparting rsistance against hot corrosion at temperature range (800-1000^{o}C) .
- Results from fuzzy model are compared with the results from experimental data . The fuzzy model predicts the weight gain with good accuracy .

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Table (1) Nominal composition of low alloy steel (Type T21-ASTM) .

Element	Fe	C	Mn	P_{max}	S_{max}	Si	Cr	Mo
Wt.%	Rem.	0.05-0.15	0.3-0.6	0.025	0.025	0.5max	2.65-3.35	0.87-1.13

Table(2) Spectrochemical analysis of low alloy steel T21.

Element	Fe	C	Mn	P	S_m	Si	Cr	Mo	V	Ti
Wt.%	Rem.	0.04	0.34	0.014	0.012	0.35	2.96	0.10	0.002	0.01

Table (3) Experimental data of hot corrosion test.

Test No.	Time (hr)	weight gain (mg/cm^2)	Weight gain (mg/cm^2)	Weight gain (mg/cm^2)
		800°C	900°C	1000°C
1	5	0.0333	0.1928	0.3333
2	10	0.0499	0.3112	0.4266
3	15	0.0603	0.3244	0.5321
4	20	0.0608	0.3679	0.6244
5	25	0.0687	0.4181	0.7048
6	30	0.0693	0.4451	0.7788
7	35	0.0811	0.4797	0.8480
8	40	0.0853	0.4811	0.9127
9	45	0.0905	0.5387	0.9740

10	50	0.0988	0.5659	1.0333
11	55	0.0999	0.5928	1.1111
12	60	0.1041	0.6166	1.1407
13	65	0.1085	0.6411	1.1999
14	70	0.1123	0.6629	1.2416
15	75	0.1166	0.7022	1.2896
16	80	0.1233	0.7058	1.3366
17	85	0.1235	0.7262	1.3815
18	90	0.1276	0.7451	1.4257
19	95	0.1661	0.7652	1.5100
20	100	0.1733	0.7839	1.5107

Table 4 values of rate constant (K_p)

Temperature (°C)	K_p (g^2 cm^{-4} hr^{-1})
800	2.4×10^{-4}
900	6×10^{-3}
1000	2.4×10^{-2}

Table 5 Crisp input values used for fuzzy logic modeling.

Input	Crisp values
Time (hr)	5-100
Temperature (°C)	800-1000

Table (6)Measured(Experimental)and Predicted data of hot corrosion test.

Test No.	Time (hr)	Measured weight gain (mg/cm^2)	Predicted weight gain (mg/cm^2)	Measured weight gain (mg/cm^2)	Predicted weight gain (mg/cm^2)	Measured weight gain (mg/cm^2)	Predicted weight gain (mg/cm^2)
		800°C	800°C	900°C	900°C	1000°C	1000°C
1	5	0.0333	0.0384	0.1928	0.1930	0.3333	0.3330
2	10	0.0499	0.0448	0.3112	0.2490	0.4266	0.5190
3	15	0.0603	0.0571	0.3244	0.3050	0.5321	0.5190
4	20	0.0608	0.0572	0.3679	0.3620	0.6244	0.6120
5	25	0.0687	0.0575	0.4181	0.4180	0.7048	0.7050
6	30	0.0693	0.0578	0.4451	0.4550	0.7788	0.7990
7	35	0.0811	0.0800	0.4797	0.4650	0.8480	0.8420

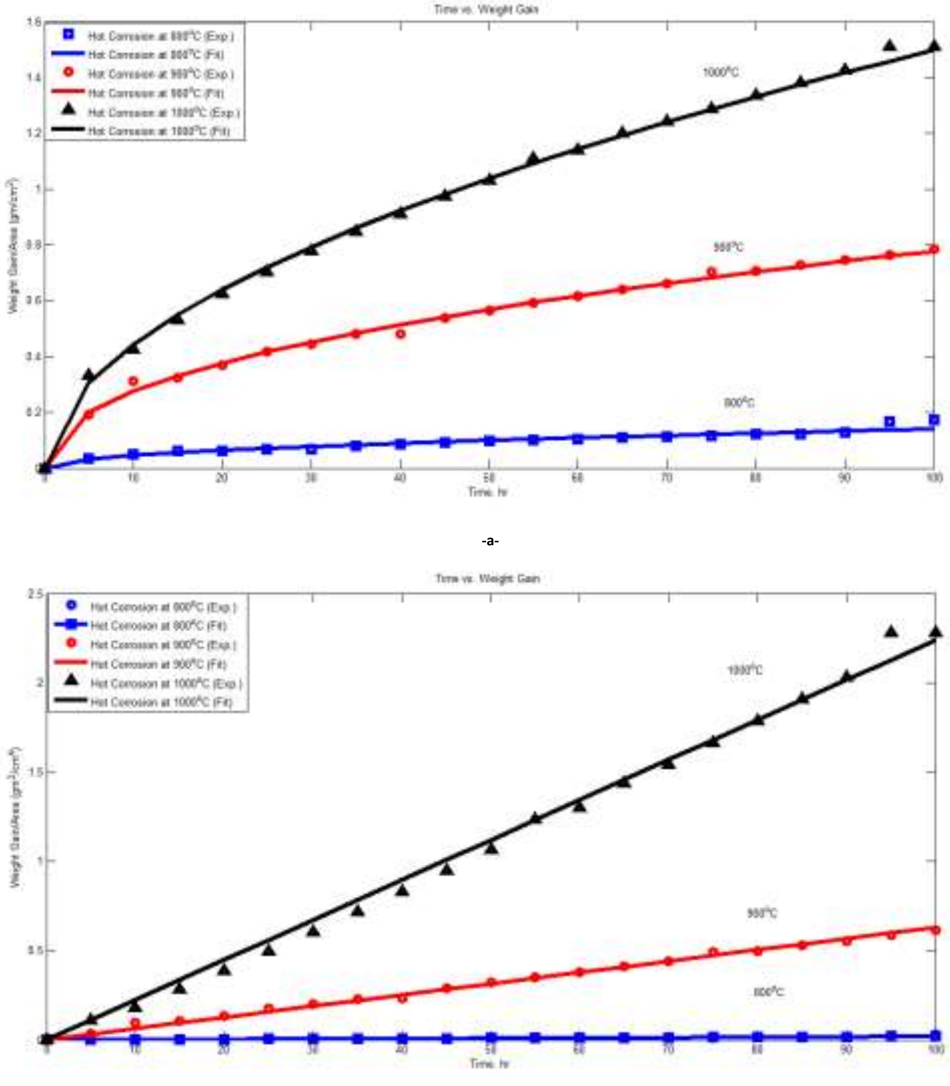
8	40	0.0853	0.0844	0.4811	0.4750	0.9127	0.9060
9	45	0.0905	0.0915	0.5387	0.5050	0.9740	0.9690
10	50	0.0988	0.0955	0.5659	0.5660	1.0333	1.0100
11	55	0.0999	0.1010	0.5928	0.5930	1.1111	1.1300
12	60	0.1041	0.1080	0.6166	0.6470	1.1407	1.1600
13	65	0.1085	0.1090	0.6411	0.6480	1.1999	1.2000
14	70	0.1123	0.1180	0.6629	0.6750	1.2416	1.2500
15	75	0.1166	0.1190	0.7022	0.6930	1.2896	1.2800
16	80	0.1233	0.1200	0.7058	0.7060	1.3366	1.3500
17	85	0.1235	0.1210	0.7262	0.7260	1.3815	1.3800
18	90	0.1276	0.1230	0.7451	0.7450	1.4257	1.4200
19	95	0.1661	0.1670	0.7652	0.7650	1.5100	1.4700
20	100	0.1733	0.1710	0.7839	0.7840	1.5107	1.5000

Table 7 Measured and predicted values of rate constant (K_p).

Temperature (°C)	K_p (g^2 cm^{-4} hr^{-1})	
	Measured	Predicted
800	2.4×10^{-4}	2×10^{-4}
900	6×10^{-3}	6.2×10^{-3}
1000	2.4×10^{-2}	2.4×10^{-2}

Table (8) Statistical Analysis (RMSE) for test cases from fuzzy logic simulation model.

Temperature (°C)	RMSE
800	0.0047
900	0.0187
1000	0.0252



-a-

-b-

Figure (1 a) weight gain/area vs time plot (parabolic fitted) for Y-doped aluminizing-siliconizing diffusion coated steel-T21 subjected to hot corrosion for 20

cycles at 100 hr for 5 cycle at temperature range (800-1000ºC) b) plot of linear-fitted for (weight gain/area)^{2}vs time.

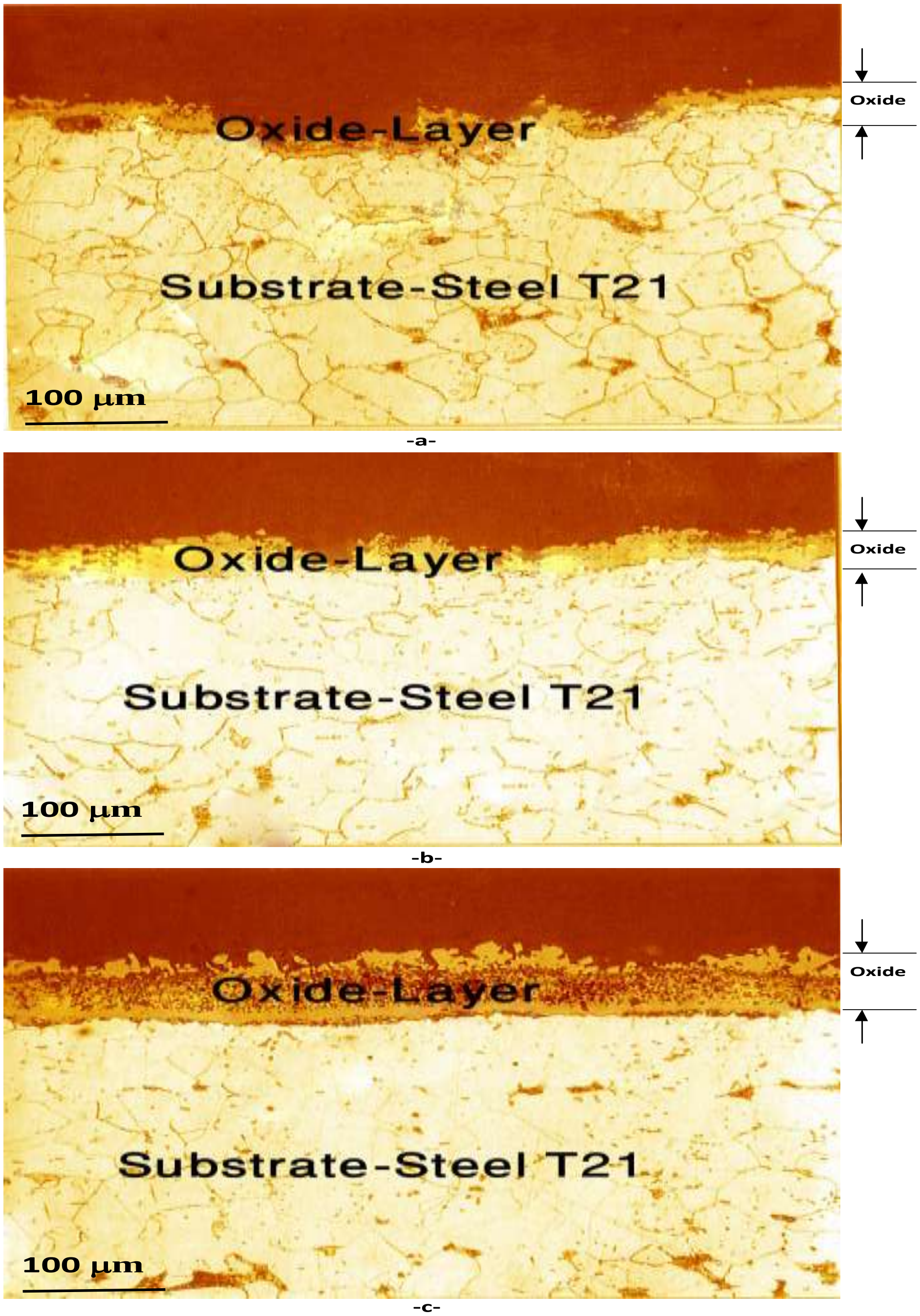


Figure (2) Light optical microscope (LOM) microstructure (Cross-Section View) of steel-T21 coated with Y-doped aluminizing-siliconizing after hot corrosion at a) 800°C and 100 hr b) 900°C and 100 hr c) 1000°C and 100 hr.

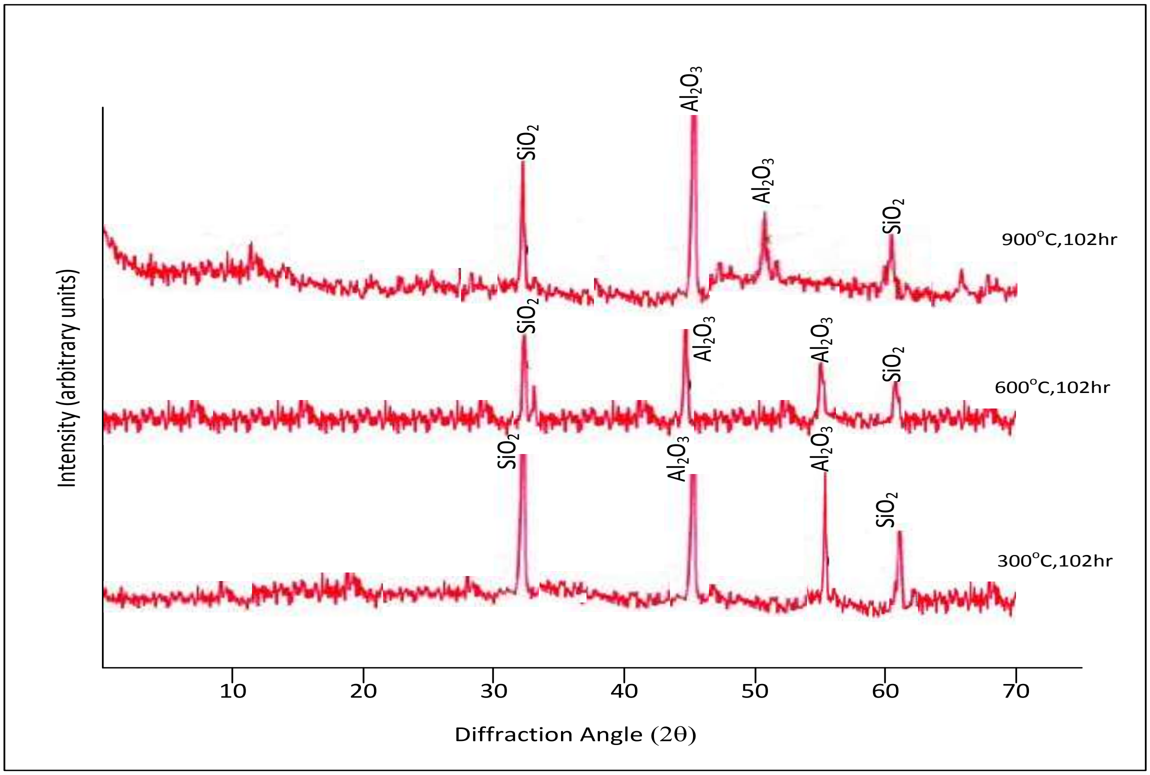


Figure (3) XRD pattern for hot corrosion tests of steel-T21 coated with Y-doped aluminizing-siliconizing.

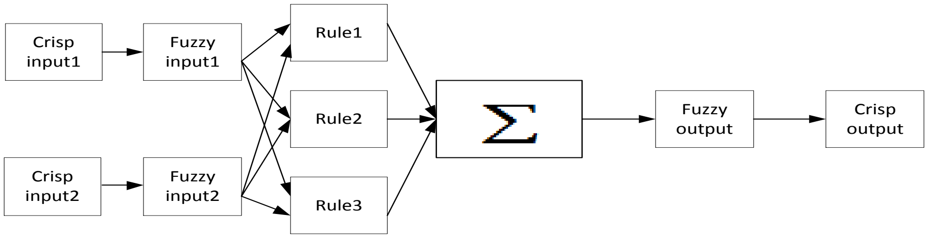
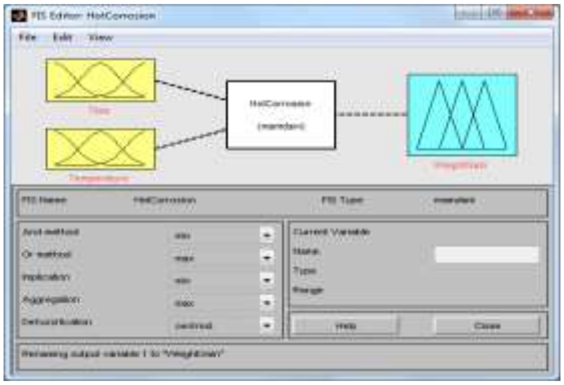
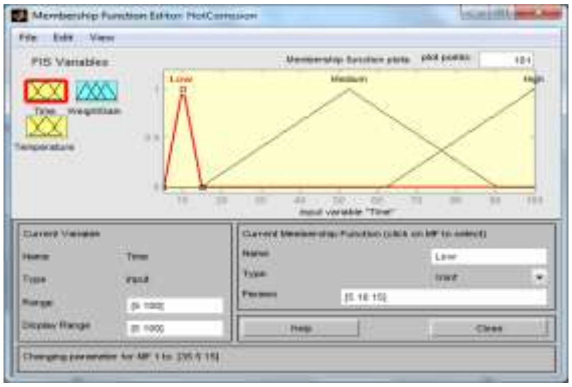


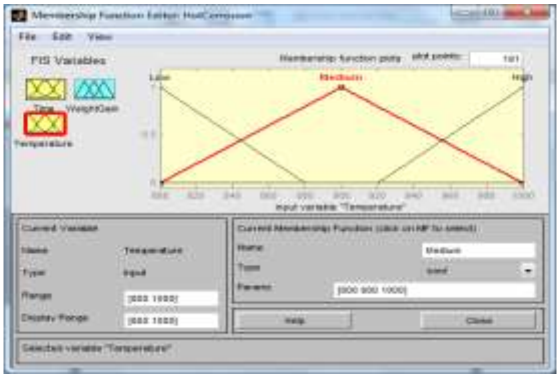
Figure (4) General structure of fuzzy interference system.



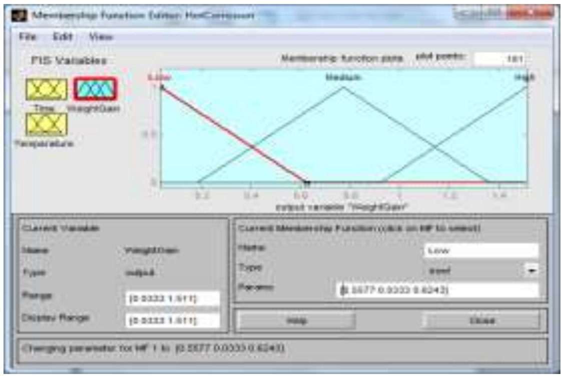
Input-output parameters for fuzzy logic control model



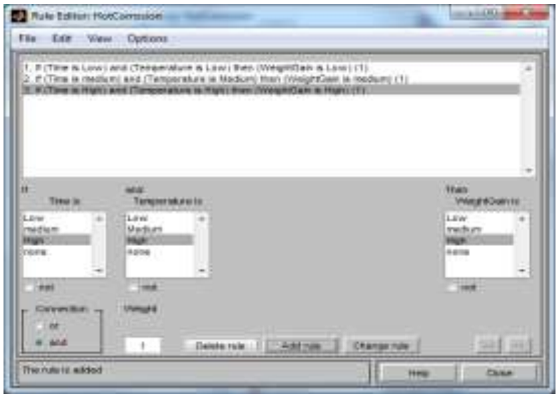
Membership function for time



Membership function for temperature

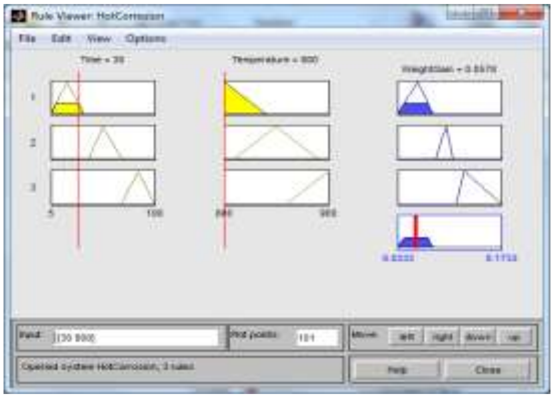


Membership function for weight gain



1. If (Time is Low) and (Temperature is Low) then (WeightGain is Low) (1)
2. If (Time is medium) and (Temperature is Medium) then (WeightGain is medium) (1)
3. If (Time is High) and (Temperature is High) then (WeightGain is High) (1)

Fuzzy rules for hot corrosion tests



Input: [30 800]

An example from fuzzy logic modeling for prediction weight gain at 30 hr, 800°C

Figure (5) Windows editor of fuzzy logic simulation stages.

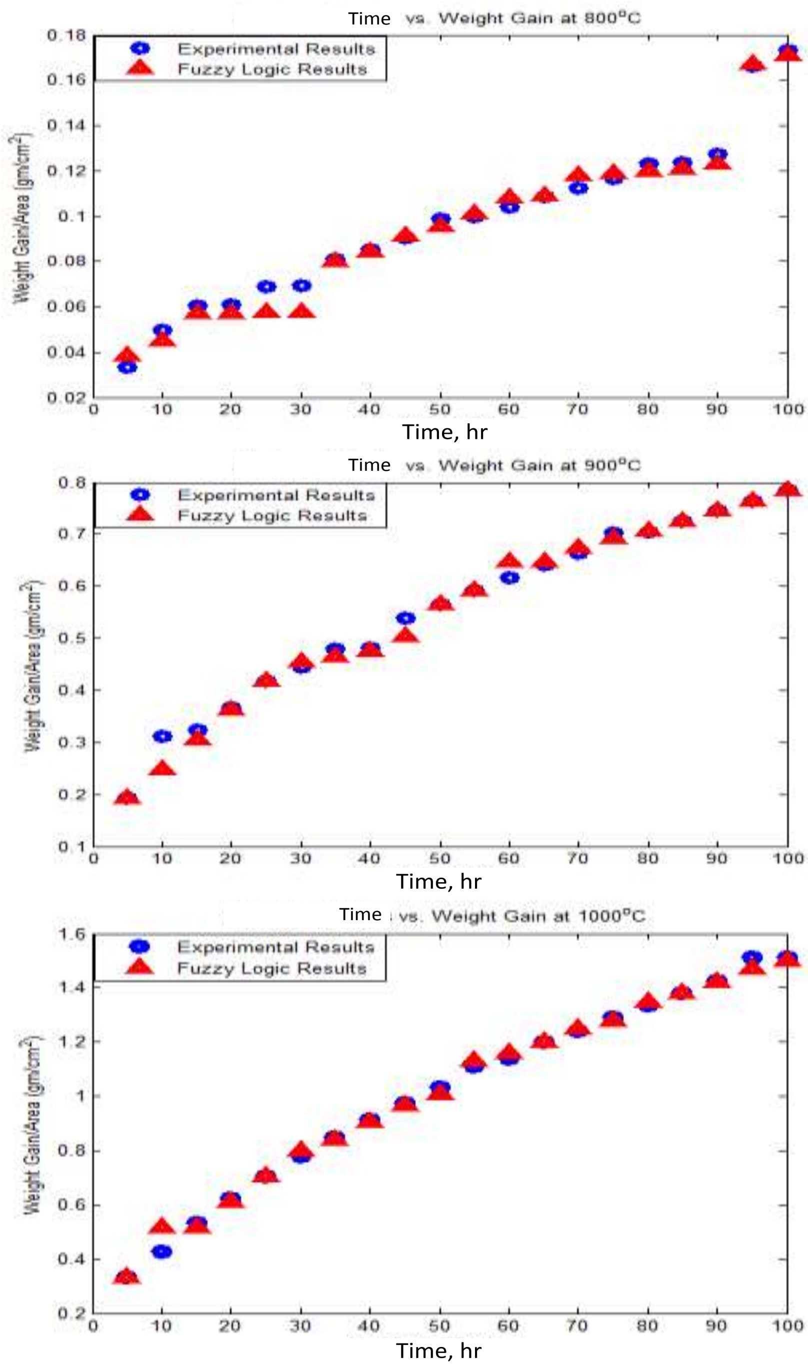


Figure (6) the relationship between the simulated (predicted) values with fuzzy logic and the measured (experimental) test results.