

## **Modeling of the Corrosion Behavior of Enamel Coatings in Different Aggressive Media**

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### **ABSTRACT**

In this study an attempt has been made to formulate materials for glass ceramic enamel coating that can be applied directly onto cast iron substrate for corrosion control. Different 'proportions(4.95) %wt. to form four batches. The batches were mixed with water to form slips which were used to coat already prepared cast iron surfaces and were allowed to dry in the oven. The coated cast iron plates were fired at temperatures of 850°C to mature and factually heat treated at 680°C.

The coatings were observed and also chemical tests were conducted to determine the corrosion properties of the resultant coatings. The chemical tests were evaluated using a suitable standard methods and special attention was paid to the type of aggressive solution, and temperature.

The results show that the coatings matured and adhered very well to the sub strateand showed good corrosion resistance in H<sub>2</sub>SO<sub>4</sub>,HCl,C<sub>6</sub>H<sub>8</sub>O<sub>7</sub>, H<sub>3</sub>PO<sub>4</sub>, and NaOH solutions. The results indicated that the corrosion rate in both acid and alkali media was increased strongly with the increasing temperature.The coatings prepared with the zircon showed better results in terms of acidcorrosion resistance than those prepared with the other crystalline agents. However, the corrosion rate of the zircon containing coating in sulfuric acid was higher than that in hydrochloric acid. Also, it is found that there is absolutely no weight loss for any of the four types of the coating in neither citric nor phosphoric acids. At the same time, the results for alkali corrosion resistance indicate that the act of the four series is very similar to each other, for the four coatings there is no high corrosion rate until the temperature exceed 100°C after that the rate increases in a very quick manner. From the results it is found that the enamel with lithium oxide was not affected by the alkali solution at all, this behavior is associated with the presence of crystalline β-Spodumene, and β-Eucryptite, which have an extremely high corrosion resistance especially for alkali medium. Mathematical modeling is implemented and regression equations are obtained by using (SPSS) software to predict the experimental data for acid and alkali corrosion rate. Comparing the predicted and measured values gives high prediction accuracy.

**Keyword:** glass-ceramic, enamel properties, SPSS software.

## نمذجة سلوك التآكل لطلاءات المينا في اوساط اكاله مختلفة

### الخلاصة:

في هذه الدراسة تم عمل محاولة لتكوين خلطة مواد مناسبة لطلاءات السيراميك الزجاجي والتي يمكن استخدامها بصورة مباشرة على الحديد الزهر للسيطرة على التآكل. تم اضافة عوامل تبلور مختلفة (CaO, Al<sub>2</sub>O<sub>3</sub>.2SiO<sub>2</sub>, ZrSiO<sub>4</sub>, TiO<sub>2</sub>, Li<sub>2</sub>O) وبنسب ثابتة (4.95%) لتكوين اربع خلطات. تم خلط هذه الخلطات مع الماء لتكوين مادة الطلاء ال(slip) والتي يتم بها طلاء اسطح حديد الزهر المحضرة ثم تترك لتجف في فرن التجفيف. بعدها الواح الحديد الزهر المطلية تحرق عند درجة 850م° للاندماج واخيرا تعامل حراريا عند درجة 680م°.

الطلاءات تم ملاحظتها وكذلك تم اجراء الفحوص الكيميائية عليها لتحديد خواص التآكل لها. تم اجراء الفحوص الكيميائية باستخدام طرق قياسية ملائمة وتم توجيه انتباه خاص لنوع المحلول الاكال المستخدم وكذلك لدرجة الحرارة. بينت النتائج ان الطلاءات قد اندمجت والتصقت بصورة جيدة جدا بالمعدن الاساس واطهرت مقاومة تاكل جيدة في المحاليل (NaOH, H<sub>3</sub>PO<sub>4</sub>, C<sub>6</sub>H<sub>8</sub>O<sub>7</sub>, HCl, H<sub>2</sub>SO<sub>4</sub>). بينت النتائج ان معدل التآكل قد ازداد بصورة كبيرة مع زيادة درجة الحرارة في كلا من الاوساط الحامضية والقاعدية. اظهرت النتائج كذلك ان الطلاءات المحضرة من الزركون لها خواص افضل من ناحية مقاومة التآكل الحامضي مقارنة مع الطلاءات الاخرى. لكن معدل التآكل للطلاءات المحتوية على الزركون في حامض الكبريتيك يكون اعلى منه في حامض الهيدروكلوريك. كذلك، وجدنا عدم حدوث أي فقدان بالوزن للطلاءات الاربعة في حامض النتريك او الفسفوريك. في نفس الوقت بينت نتائج مقاومة التآكل في المحاليل القاعدية ان السلوك كان متشابه جدا للطلاءات الاربعة، حيث لم يلاحظ أي معدل تاكل عالي حتى درجة 100م° بعدها ازداد معدل التآكل بصورة سريعة جدا. وقد وجدنا من خلال النتائج ان الطلاء الحاوي على اوكسيد الليثيوم لم يتاثر بالمحلول القاعدي بصورة تامة، وهذا نتيجة وجود الاطوار البلورية (β-Eucryptite، وβ-Spodumene) والتي لها مقاومة تاكل عالية جدا وخصوصا للاوساط القاعدية. تم أيضا استخدام موديل رياضي و تم الحصول على معادلات انحدار باستخدام برنامج (SPSS) والتي تكونت لغرض التنبؤ بالبيانات المختبرية لخواص الطلاء. بينت النتائج من خلال المقارنة بين القيم المتنبئة والحقيقية ان هذا البرنامج يعطي دقة تنبؤ عالية.

## INTRODUCTION

The problem of metallic corrosion is one of significant proportions to any nation. In economic terms it has been estimated that approximately 5% of an industrialized nation's income is spent on prevention and the replacement of products lost or contaminated as a result of corrosion reactions [1].

Cast iron is a generic term that identifies a large family of ferrous alloys. Low raw material costs and relative ease of manufacture make cast irons the least expensive of the engineering metals. Cast irons may often be used in place of steel at considerable cost savings. These materials are used in the food, brewery, chemical processing, thermal power plants, metallurgical and other industries. Equipment and structures in these industries placed in areas near the sea deteriorate rapidly if proper protective schemes are not employed to protect them. Presently the principal method used for controlling corrosion in some of the industries includes the use of organic paints, which do not give lasting protection. In this case the equipment is repainted after a short time at a great cost. Also some of the industries simply replace machine parts at a great cost due to corrosion damage. Some advertising bill boards have also been found to corrode which often become an eyesore and a hazard and are therefore replaced after a short time [2].

Protecting these ferrous substrates mentioned above with good coating is therefore of utmost importance. The use of inorganic coatings such as enamels or properly fused

ceramic coatings on these surfaces can produce a more lasting solution to the problem of corrosion as these coatings are chemically inert and also possess superior mechanical properties. They protect by acting as a barrier between the environment and the metal surface and thus substantially minimize the corrosion process. Also because of their chemical inertness they do not pose any danger to the environment and are therefore considered to be environmentally friendly.

Enamel is inorganic coatings based on a special ceramic vitreous matrix in which specific additives are randomly dispersed. The ceramic vitreous matrix is made by a mixture of various raw materials and elements and in particular it is based on boron silicate glass added with metal oxides of zinc, sodium, aluminum, etc. these addition are often used to improve and enhance some important performances such as corrosion and wear resistance, and also aesthetic functions [3].

A great deal of effort was devoted to the improvement of the mechanical and chemical properties of enamel coatings in the last few decades aiming at enlarging their field of applications. Approaches to improve the corrosion resistance of enamel include the incorporation of solid particles, such as quartz [4-5], zirconia [6], or by using double layer of enamel [7].

The main objective of this project is to formulate an appropriate corrosion resistance inorganic (glass-ceramic enamel) coating that can be used for corrosion control on cast iron substrates. The present paper describes the preparation, process of application, and evaluation of this developed coating. Mathematical model and investigation of the effects of crystallization agents ( $\text{Li}_2\text{O}$ ,  $\text{TiO}_2$ ,  $\text{ZrSiO}_4$ , and  $\text{CaO} \cdot \text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$ ) on the properties related to corrosion resistance of coating have been also reported in this paper. Different corrosive media ( $\text{H}_2\text{SO}_4$ ,  $\text{HCl}$ ,  $\text{C}_6\text{H}_8\text{O}_7$ ,  $\text{H}_3\text{PO}_4$ , and  $\text{NaOH}$ ), various pH and temperatures were used.

### Experimental

A sheet of cast iron (3.6 % C) was used as the enamel substrate (the chemical composition of selected cast iron are listed in Table (1)) and exposed to a jet of an abrasive material (quartz) shots to remove scale, rust and dirt. The surface becomes clean and slightly pitted which helps promote good bonding. Removal of material from the surface should not be excessive and is controlled by adjusting the air pressure and exposure time of blasting.

**Table (1) chemical composition of metal substrates**

Element	C	Mn	P	S	Si	Fe
Cast Iron	3.6	0.5	0.65	0.85	2.25	92.15

Before blasting, oil and drawing compounds are removed by heating the surface at  $455^\circ\text{C}$  to burn off the organic contaminants.

The reagents of the enamels were mainly composed of  $\text{SiO}_2$ ,  $\text{CaO} \cdot \text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$ ,  $\text{Na}_2\text{B}_4\text{O}_7 \cdot 10\text{H}_2\text{O}$ ,  $\text{ZnO}$ ,  $\text{MnO}$ ,  $\text{KNO}_3$ ,  $\text{CoO}$ ,  $\text{NiO}$ , and carbonates of alkali metals and alkaline earth metals. The batch formula of the enamels was listed in Table 2, where crystallization agents ( $\text{Li}_2\text{O}$ ,  $\text{TiO}_2$ ,  $\text{ZrSiO}_4$ , and  $\text{CaO} \cdot \text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$ ) were added in an appropriate weight ratio to prepare four different batches of enamel coatings and to study their effect on corrosion properties of resulting coatings. The reagents of 20 g weight were pulverized, mixed, and melted in a graphite crucible, using a high temperature

electric resistance furnace, at 1250°C for 2 hr. The molten enamels were stirred with a ceramic rod, and quenching into water to produce glassy powder called frit, which is the main coating material. The as quenched frits of each of the four batches of the enamel coating were ball milled separately down to 200 mesh and mixed, with equal part of each frit and additive of kaolinite, quartz, borax, bintonite, and water, to form a batch of enamel slip. The specific gravity of the enameling slip was controlled between 1.6-1.7 by adjusting the water content, and the slip was aged for 24hr before enameling to improve its fluidity. The enamel slip was then applied to the pretreated sheet of cast iron by a dipping method. The specimens were then dried in an oven at 150°C for 30min. firing of the enamel coating was carried out in a box furnace at 820°C for 4min and cooled in air. Subsequently, the coatings were heat-treated at 680°C for 1 h. The entire process of glass-ceramic coating is summarized in the flow diagram of experimental program as shown in Figure (1).

**Table (2) Weight percent of four types of frit.**

Material	C1	C2	C3	C4
BaCO <sub>3</sub>	9.24	9.24	9.24	9.24
Co <sub>3</sub> O <sub>4</sub>	1.43	1.43	1.43	1.43
Na <sub>2</sub> O.2B <sub>2</sub> O <sub>3</sub> .2SiO <sub>2</sub>	20.16	20.16	20.16	20.16
CaO.Al <sub>2</sub> O <sub>3</sub> .2SiO <sub>2</sub>	16.74	16.74	16.74	21.69
KNO <sub>3</sub>	8.48	8.48	8.48	8.48
Li <sub>2</sub> O	4.95	-----	-----	-----
MnO	1.25	1.25	1.25	1.25
NiO	2.32	2.32	2.32	2.32
SiO <sub>2</sub>	33.74	33.74	33.74	33.74
TiO <sub>2</sub>	-----	-----	4.95	-----
ZnO	1.69	1.69	1.69	1.69
ZrSiO <sub>4</sub>	-----	4.95	-----	-----

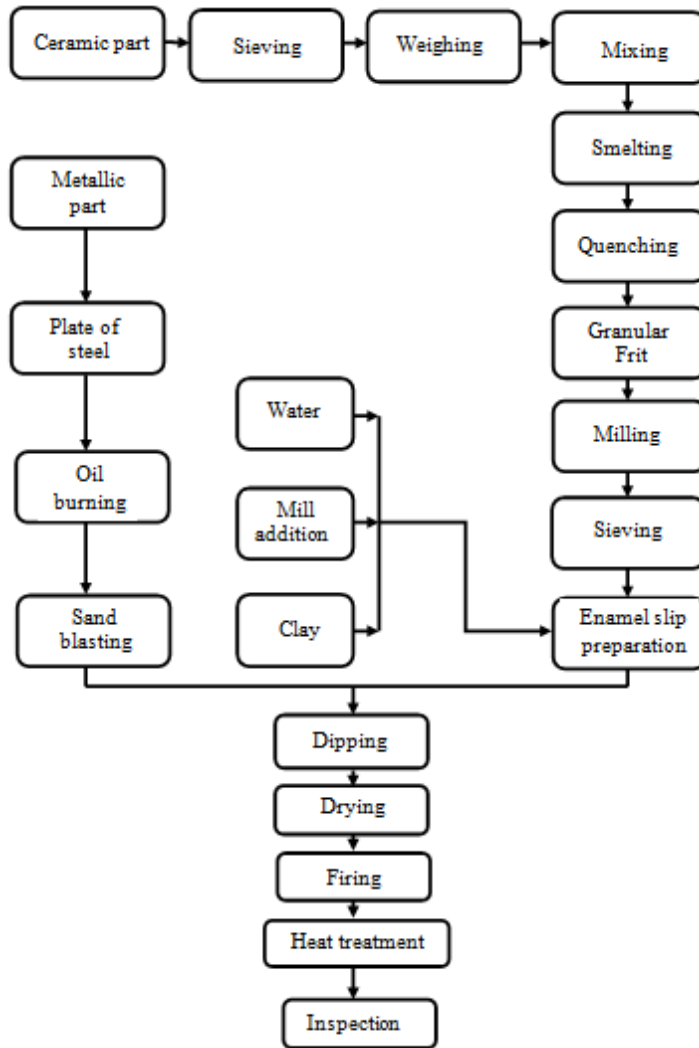


Figure (1) Flow diagram of experimental program

**Coating Characterization**

The thickness of the resultant coating was measured by an eddy current based thickness measuring instrument with ND-2 type probe, suitable for non-ferrous alloys. Phase analysis of the resultant coating before or after heat treatment was done by X-ray diffraction analysis (Philips PLO1840 X-ray diffract to meter in  $2\theta$  ranging between  $10^\circ$  to  $90^\circ$  using Cu  $K\alpha$  radiation) the results of this test were discussed in previous paper [8].

**Corrosion tests**

A set of tests for acid and alkali resistance for coatings were made. Special attention was given to temperature, and pH. The tests were made at constant time 9 hr. with

temperature ranges from (208-463)°K the increasing rate was 25°C. These tests were performed according to the ASTM C703.

In every case the corrosion rate was determined by weight loss and calculated using equation below [9]:

$$\text{Corrosion Rate (mdd)} = \Delta w / (A \cdot t) \quad \dots(1)$$

Where:

**mdd:** mg/day.dcm<sup>2</sup>, **Δw:** weight loss (gm), **A:** surface area (dcm<sup>2</sup>), **t:** time (day).

In mill per year corrosion rate is calculated from equation [5]:

$$C.R(\text{mpy}) = 1.44 * mdd / S.G \quad \dots(2)$$

*S.G:* Specific gravity.

**Mathematical modeling**

A statistical model for the prediction of the corrosion rate of the resultant coating was created by regression function in SPSS software from the training data set. The definition of the variation factors (independent variables) and their values are given in Table 3, while the dependent variables (response functions) were corrosion rate at hydrochloric acid (H), corrosion rate at sulfuric acid (S), and corrosion rate at sodium hydroxide (N).

**Table (3) Definition and values of independent variables used in regression equation**

Designations of independent variable	Name of variable	Value	Interval
	Temperature (°C)	25 - 200	25
	Zircon sand concentration (%)	16.74 - 21.69	4.95
	Lithium oxide concentration (%)	0 - 4.95	4.95
	Titanium oxide concentration (%)	0 - 4.95	4.95

All original 36 samples within experimental data shown in Table 4 were randomly divided into two data sets; the training data set and the testing data set. The training data set contained 24 samples which were used to build a prediction model as shown in Table 5 and the testing data set contained 12 samples which were used to test the flexibility and the validity of the prediction model as shown in Table (6).

Table (4) Original experimental data for predicted and measured corrosion tests (mpy).

No	Temperature	Zircon sand addition	Feldspar addition	Lithium oxide addition	Titanium oxide addition	Measured corrosion rate (mpy) in HCl	Predicted corrosion rate (mpy) in HCl	Measured corrosion rate (mpy) in H <sub>2</sub> SO <sub>4</sub>	Predicted corrosion rate (mpy) in H <sub>2</sub> SO <sub>4</sub>	Measured corrosion rate (mpy) in NaOH	Predicted corrosion rate (mpy) in NaOH
1	.00	4.95	16.74	0	0	0	-0.61906	0	-0.53844	0	-0.24228
2	25	4.95	16.74	0	0	0	-0.22818	0	-0.12328	0	-0.10365
3	50	4.95	16.74	0	0	0	0.16269	0	0.29189	0	0.03497
4	75	4.95	16.74	0	0	0	0.55357	0.1	0.70706	0	0.1736
5	100	4.95	16.74	0	0	0.6	0.94444	1.1	1.12222	0	0.31222
6	125	4.95	16.74	0	0	1.2	1.33532	1.6	1.53739	0.05	0.45085
7	150	4.95	16.74	0	0	1.6	1.72619	2.1	1.95256	0.06	0.58947
8	175	4.95	16.74	0	0	2.3	2.11707	2.4	2.36772	1.2	0.7281
9	200	4.95	16.74	0	0	2.8	2.50794	2.8	2.78289	1.5	0.86672
10	.00	0	21.69	0	0	0	-0.59683	0	-0.47956	0	-0.16561
11	25	0	21.69	0	0	0	-0.20596	0	-0.06439	0	-0.02699
12	50	0	21.69	0	0	0	0.18492	0	0.35078	0	0.11164
13	75	0	21.69	0	0	0	0.57579	0.1	0.76594	0	0.25026
14	100	0	21.69	0	0	0.7	0.96667	1.2	1.18111	0	0.38889
15	125	0	21.69	0	0	1.2	1.35754	1.8	1.59628	0.05	0.52751
16	150	0	21.69	0	0	1.7	1.74842	2.1	2.01144	0.06	0.66614
17	175	0	21.69	0	0	2.2	2.13929	2.6	2.42661	1.49	0.80476
18	200	0	21.69	0	0	2.9	2.53017	2.83	2.84178	1.9	0.94339
19	.00	0	16.74	4.95	0	0	-0.54683	0	-0.504	0	-0.5545
20	25	0	16.74	4.95	0	0	-0.15596	0	-0.08883	0	-0.41588
21	50	0	16.74	4.95	0	0	0.23492	0	0.32633	0	-0.27725
22	75	0	16.74	4.95	0	0	0.62579	0.1	0.7415	0	-0.13863
23	100	0	16.74	4.95	0	0.8	1.01667	1.3	1.15667	0	0
24	125	0	16.74	4.95	0	1.25	1.40754	1.81	1.57183	0	0.13863
25	150	0	16.74	4.95	0	1.8	1.79842	2	1.987	0	0.27725
26	175	0	16.74	4.95	0	2.4	2.18929	2.4	2.40217	0	0.41588
27	200	0	16.74	4.95	0	2.9	2.58017	2.8	2.81733	0	0.5545
28	.00	0	16.74	0	4.95	0	-0.50461	0	-0.41733	0	-0.18672
29	25	0	16.74	0	4.95	0	-0.11374	0	-0.00217	0	-0.0481
30	50	0	16.74	0	4.95	0	0.27714	0	0.413	0	0.09053
31	75	0	16.74	0	4.95	0	0.66801	0.1	0.82817	0	0.22915
32	100	0	16.74	0	4.95	0.8	1.05889	1.28	1.24333	0	0.36778
33	125	0	16.74	0	4.95	1.3	1.44976	1.81	1.6585	0.06	0.5064
34	150	0	16.74	0	4.95	1.83	1.84064	2.3	2.07367	0.25	0.64503
35	175	0	16.74	0	4.95	2.5	2.23151	2.7	2.48883	1.3	0.78365
36	200	0	16.74	0	4.95	3.1	2.62239	3	2.904	1.7	0.92228

Table (5) Training data for corrosion tests (mpy).

No	Temperature	Zircon sand addition	Feldspar addition	Lithium oxide addition	Titanium oxide addition	Measured corrosion rate (mpy) inHCl	Predicted corrosion rate (mpy) inHCl	Measured corrosion rate (mpy) in H <sub>2</sub> SO <sub>4</sub>	Predicted corrosion rate (mpy) in H <sub>2</sub> SO <sub>4</sub>	Measured corrosion rate (mpy) in NaOH	Predicted corrosion rate (mpy) in NaOH
1	.00	4.95	16.74	0	0	0	-0.61906	0	-0.53844	0	-0.24228
2	25	4.95	16.74	0	0	0	-0.22818	0	-0.12328	0	-0.10365
3	50	4.95	16.74	0	0	0	0.16269	0	0.29189	0	0.03497
4	75	4.95	16.74	0	0	0	0.55357	0.1	0.70706	0	0.1736
5	100	4.95	16.74	0	0	0.6	0.94444	1.1	1.12222	0	0.31222
6	125	4.95	16.74	0	0	1.2	1.33532	1.6	1.53739	0.05	0.45085
10	.00	0	21.69	0	0	0	-0.59683	0	-0.47956	0	-0.16561
14	100	0	21.69	0	0	0.7	0.96667	1.2	1.18111	0	0.38889
15	125	0	21.69	0	0	1.2	1.35754	1.8	1.59628	0.05	0.52751
16	150	0	21.69	0	0	1.7	1.74842	2.1	2.01144	0.06	0.66614
17	175	0	21.69	0	0	2.2	2.13929	2.6	2.42661	1.49	0.80476
18	200	0	21.69	0	0	2.9	2.53017	2.83	2.84178	1.9	0.94339
19	.00	0	16.74	4.95	0	0	-0.54683	0	-0.504	0	-0.5545
21	50	0	16.74	4.95	0	0	0.23492	0	0.32633	0	-0.27725
22	75	0	16.74	4.95	0	0	0.62579	0.1	0.7415	0	-0.13863
25	150	0	16.74	4.95	0	1.8	1.79842	2	1.987	0	0.27725
26	175	0	16.74	4.95	0	2.4	2.18929	2.4	2.40217	0	0.41588
27	200	0	16.74	4.95	0	2.9	2.58017	2.8	2.81733	0	0.5545
28	.00	0	16.74	0	4.95	0	-0.50461	0	-0.41733	0	-0.18672
29	25	0	16.74	0	4.95	0	-0.11374	0	-0.00217	0	-0.0481
30	50	0	16.74	0	4.95	0	0.27714	0	0.413	0	0.09053
31	75	0	16.74	0	4.95	0	0.66801	0.1	0.82817	0	0.22915
32	100	0	16.74	0	4.95	0.8	1.05889	1.28	1.24333	0	0.36778
36	200	0	16.74	0	4.95	3.1	2.62239	3	2.904	1.7	0.92228



Table (6) testing data for corrosion tests (mpy)

No	Temperature	Zircon sand addition	Feldspar addition	Lithium oxide addition	Titanium oxide addition	Measured corrosion rate (mpy) in HCl	Predicted corrosion rate (mpy) in HCl	Measured corrosion rate (mpy) in H <sub>2</sub> SO <sub>4</sub>	Predicted corrosion rate (mpy) in H <sub>2</sub> SO <sub>4</sub>	Measured corrosion rate (mpy) in NaOH	Predicted corrosion rate (mpy) in NaOH
7	150	4.95	16.74	0	0	1.6	1.72619	2.1	1.95256	0.06	0.58947
8	175	4.95	16.74	0	0	2.3	2.11707	2.4	2.36772	1.2	0.7281
9	200	4.95	16.74	0	0	2.8	2.50794	2.8	2.78289	1.5	0.86672
11	25	0	21.69	0	0	0	-0.20596	0	-0.06439	0	-0.02699
12	50	0	21.69	0	0	0	0.18492	0	0.35078	0	0.11164
13	75	0	21.69	0	0	0	0.57579	0.1	0.76594	0	0.25026
20	25	0	16.74	4.95	0	0	-0.15596	0	-0.08883	0	-0.41588
23	100	0	16.74	4.95	0	0.8	1.01667	1.3	1.15667	0	0
24	125	0	16.74	4.95	0	1.25	1.40754	1.81	1.57183	0	0.13863
33	125	0	16.74	0	4.95	1.3	1.44976	1.81	1.6585	0.06	0.5064
34	150	0	16.74	0	4.95	1.83	1.84064	2.3	2.07367	0.25	0.64503
35	175	0	16.74	0	4.95	2.5	2.23151	2.7	2.48883	1.3	0.78365

## Results and discussion

### Corrosion tests

#### Acid corrosion rate

Four different types of acids (H<sub>2</sub>SO<sub>4</sub>, HCl, C<sub>6</sub>H<sub>8</sub>O<sub>7</sub>, and H<sub>3</sub>PO<sub>4</sub>) with pH=2 were used, and tests have been performed at temperature range 298 to 463 K (25-125) °C according to ASTM C703 with 9 hr. of immersing in acidic solutions. The results are present in Figs 2 (a & b). From results it is found that there is absolutely no weight loss for any of the four types of frits in neither citric nor phosphoric acids, and there is no weight loss at room temperature at all of the used corrosive media. Also, the results show that the H<sub>2</sub>SO<sub>4</sub> acid has more aggressive effect than HCl acid. Corrosion rate increases largely with increasing the temperature.

Corrosion resistance of zircon containing enamel is very high comparing with the other types of crystallization agents this is to the formation of ionic species like ZrO<sup>+3</sup>, Zr<sup>+4</sup>, and HZrO<sup>-3</sup> will only occur below pH~0 and above pH~17, respectively. Thus the hydrated ZrO<sub>2</sub> surface is stable at all the conceivable pH values of the solution and probably offers a very high activation barrier for diffusion of the ionic species through it [10]. These results agree well with those obtained by Nagham S. Hassan [6]. Also, titania containing enamel shows high corrosion resistance, this is because TiO<sub>2</sub> is thermodynamically stable in aqueous solutions, indeed the addition of it to a glass increases its chemical durability [11]. Lithia and feldspar containing enamels have very close corrosion resistance but lower than that of zircon and titania enamels.

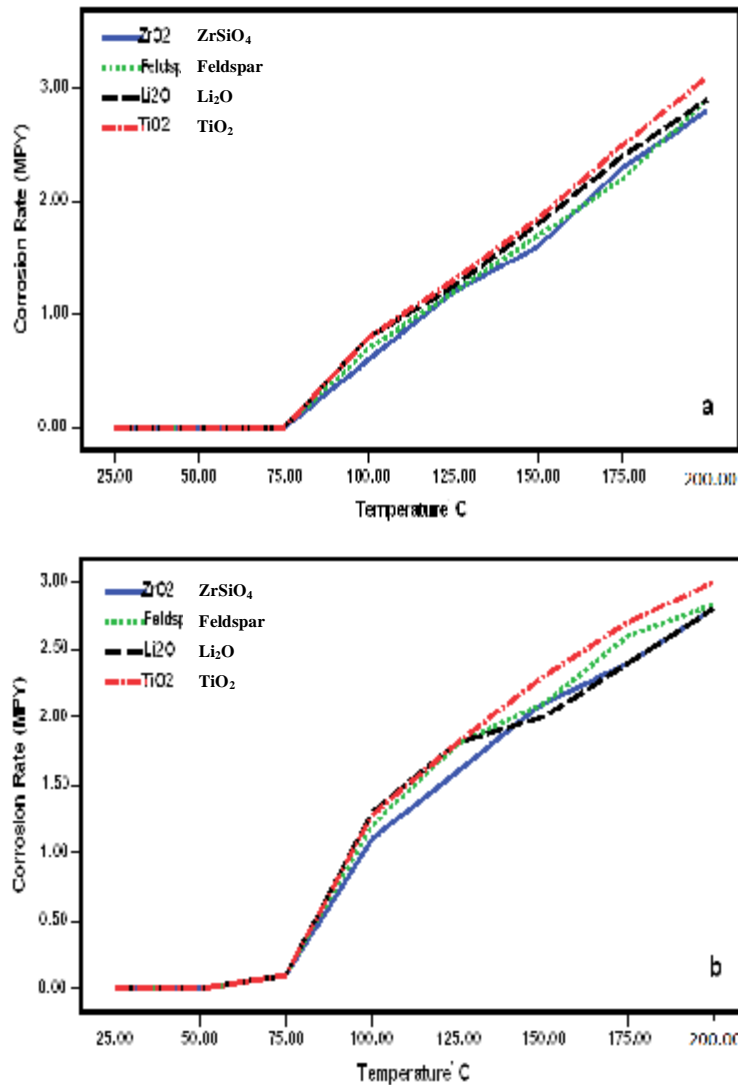


Figure (2) Corrosion rate Vs. Temperature of enameled cast iron in (a) HCl, (b) H<sub>2</sub>SO<sub>4</sub> (pH=2) solution

**Alkali corrosion rate**

Using of enamel as a protecting surface against the action of alkali solutions is wide spread, especially in washing machines where high concentration of alkali is usual. All the enamels in this study have not been affected by an alkali solution of NaOH with pH=12 at room temperature.

The standard test for alkali resistance is ASTM C614, which indicates the resistance of a enamel to a hot alkali solution. Alkali resistance is expressed as MPY in terms of weight loss for the area exposed to the test solution. According to this ASTM the corrosion resistance of enameled cast iron to NaOH (pH=12) solution at high temperatures is shown in Fig 3. Corrosion rate has increased largely with the

increasing of the temperature. Here the act of the four series is very similar to each other, for the four enamels there is no high corrosion rate until the temperature exceed 100°C after that the rate increases in a very quick manner. From the figure it has seen that the enamel with lithium oxide was not affected by the alkali solution at all, this behavior is associated with the presence of crystalline β-Spodumene, and β-Eucryptite, which have an extremely high corrosion resistance especially for alkali medium [12, 13]. Zircon sand enamel has higher corrosion resistance than titania and feldspar containing enamel.

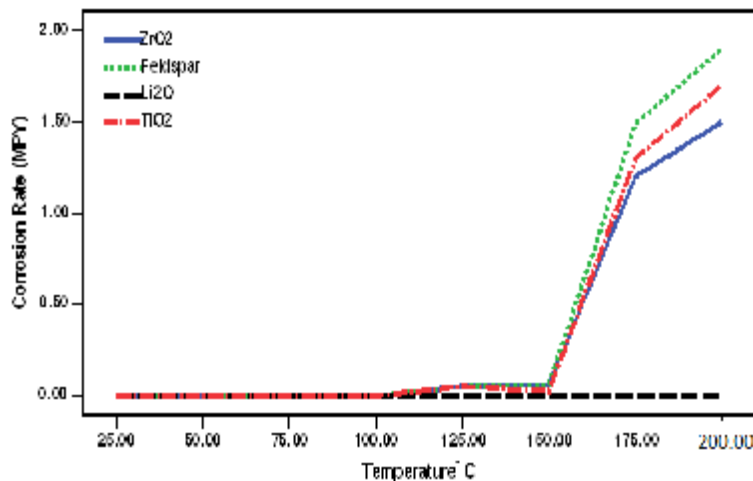


Figure (3) Corrosion rate Vs. Temperature of enameled cast iron in NaOH (pH=12)

**Mathematical model**

After processing of experimental results, mathematical models(regression equation) for corrosion rates of coating were obtained:-

$$H = -0.597 + 0.016X_1 - 0.004X_2 + 0.01X_3 + 0.019X_4$$

$$S = -0.48 + 0.017X_1 - 0.012X_2 - 0.005X_3 + 0.013X_4$$

$$N = -0.166 + 0.006X_1 - 0.015X_2 - 0.079X_3 - 0.004X_4$$

Where:

H: corrosion rate(mpy) at hydrochloric acid, S : corrosion rate(mpy) at sulfuric acid, N : corrosion rate(mpy) at sodium hydroxide, X<sub>1</sub>: temperature (°C), X<sub>2</sub>: zircon concentration (%), X<sub>3</sub>: lithium oxide concentration (%), X<sub>4</sub>: titanium oxide concentration (%).

The values of the multiple correlation coefficient R, that tell us how strongly the multiple independent variables are related to the dependent variable, were (0.947, 0.960, & 0.756) for equations (3, 4, & 5) respectively.

The result of mathematical modeling shows that the feldspar content has a little effect comparing with other factors. Therefore, the feldspar term was negligible from equations.

The negative values of corrosion rate obtained from equations are imaginary values; because there is no corrosion rate in negative value. Therefore, these values indicate there is no corrosion situation.

Figures (4-6) show the comparison between the predicted values and measured values of 28 original data for acid corrosion rate and alkali corrosion rate respectively by using (SPSS) software.

It is clear from these figures that the predicted values are in a close match with the measured values for all properties.

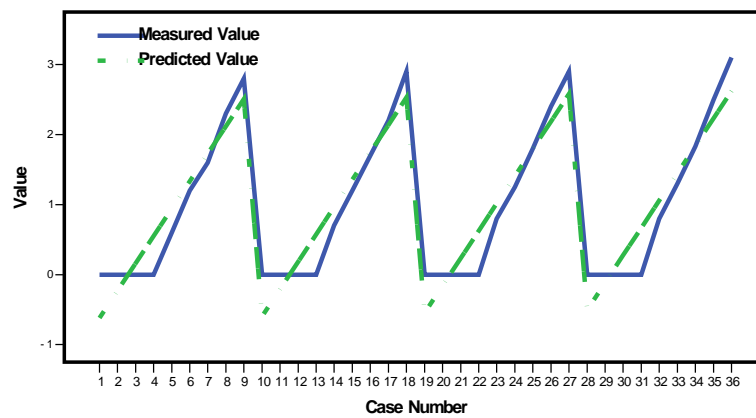


Figure (4) Comparison between Measured and Predicted values for the experimental data for corrosion rate at HCl.

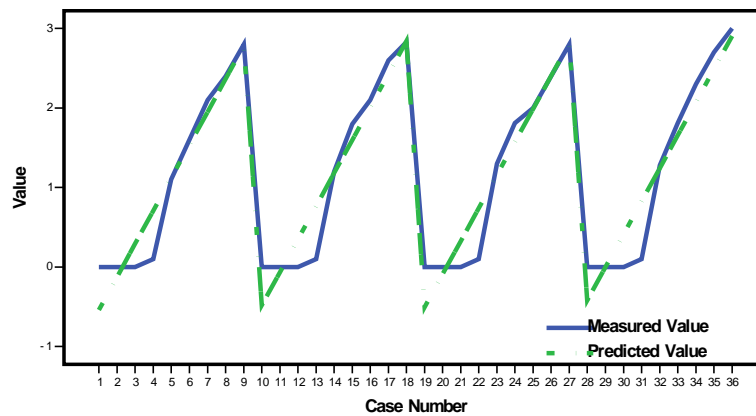
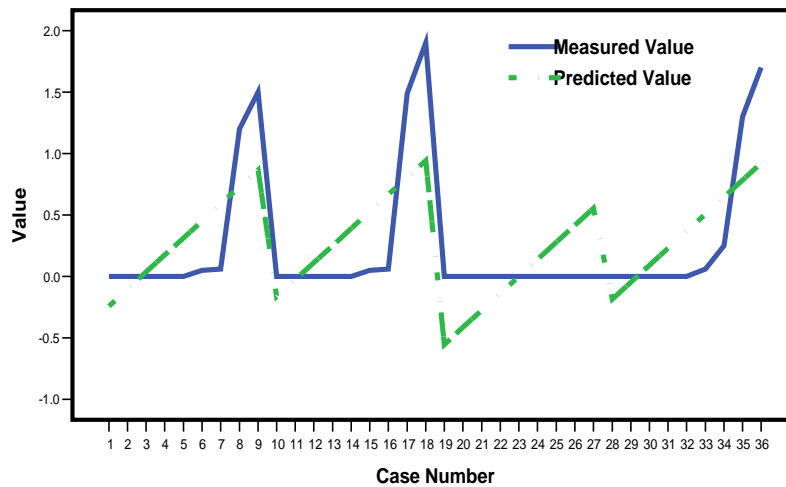


Figure (5) Comparison between Measured and Predicted values for the experimental data for corrosion rate at H<sub>2</sub>SO<sub>4</sub>.



**Figure (6) Comparison between Measured and Predicted values for the experimental data for corrosion rate at NaOH**

## CONCLUSION

The following conclusions may be drawn from the results obtained in this work:

1. The coating material can be applied as a single coat by simple vitreous enameling process on selected cast iron alloy.
2. Added the crystalline agent increases the corrosion resistance of the resultant coatings in all cases.
3. The coatings prepared with the zircon showed better results in terms of acid corrosion resistance than those prepared with the other crystalline agents, while the coatings with lithium oxide have not affected by the alkali solution at all.
4. The proposed ceramic coating material possesses reasonable low melting temperature (1200-1250°C) and processing 860°C temperature, thus leading to less cost and energy consumption during preparation and application.
5. The formation of different crystalline phases in resultant coating depends on initial composition, and the type of the added crystalline agent.
6. The multiple regression models could predict the acid and alkali resistance properties with higher accuracy for different crystalline agent addition, and temperature.

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