

## Studying and Modeling The Effects of Quartz Addition and Heat Treatment on Corrosion Properties of Ceramic Coating

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### Abstract

In this work, a new glass-ceramic coating has been developed and applied, as a single coat without prior chemical treatment of the surface, by using the dipping technique on metal substrate. the coating are designed for application on varies grades of low alloy steel, the selected substrate was low carbon low alloyed steel with (0.2)%C. Various heat treatments at temperatures ( 500,550, and 600°C ) at different times (60 & 120)min and with quartz addition in the range (0-15)% were used to obtain a glass-ceramics that have the optimum coating properties. These coating have been characterized by x-ray diffraction analyses and the results showed presence of a number of microcrystalline phases which are formed during the heat treatments. The results of corrosion resistance tests indicated that the acid resistance was greatly improved by addition of quartz into enamel frit but at same times decreased the alkali corrosion resistance, while the heat treatments improved both acid and alkali resistance for all cases, this is attributed to the formation of complex network from crystalline phases. The results also indicated that the heat treatment at 600°C for 120min with 15% quartz addition brought the optimum values for acid resistance property which are improved by 86.66%, while the optimum values for alkali resistance property was brought with free quartz added samples which were treated at 600°C for 120min. 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. The accuracy of prediction has been (82.58%, and 96.7%) for acid and alkali resistance properties respectively.

**Keywords:** glass-ceramic, corrosion, SPSS software

### دراسة ونمذجة تأثير إضافات الكوارتز والمعاملة الحرارية على خواص التآكل للطلاء السيراميكي

#### الخلاصة

في هذا البحث، تم تطوير طلاء سيراميك زجاجي جديد، وتمت عملية الطلاء بدون معاملة كيميائية مسبقة للسطح، باستخدام تقنية التغطيس على المعدن الأساس. وتم تصميم الطلاء لاستخدامه لكافة أنواع الفولاذ ألسبائكي. تم إجراء التعامل الحراري عند درجات حرارية ( 500, 550, 600 ) درجة مئوية وعند فترات زمنية مختلفة ( 60 & 120 ) دقيقة مع إضافة كوارتز بنسبة تتراوح بين ( 0-15 )% للحصول على سيراميك زجاجي ذو خواص طلاء عالية .

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وتم التعرف على الأطوار المتبلورة باستخدام التحليل بالأشعة السينية وبينت النتائج وجود عدد من الأطوار المتبلورة والتي تكونت خلال المعاملة الحرارية. أظهرت النتائج أن أضافت الكوارتز تحسن بصورة كبيرة مقاومة الحوامض لكن في نفس الوقت تقلل من مقاومة القواعد، بينما المعاملة الحرارية تؤدي إلى تحسين كلا من مقاومة الحوامض والقواعد في جميع الحالات التي تم دراستها. وذلك بسبب تكون شبكة من الأطوار المتبلورة المعقدة والتي تؤدي إلى زيادة مقاومة التآكل في الأوساط الحامضة والقاعدية.

بينت النتائج أيضا انه بالتعامل الحراري عند 600 درجة مئوية لمدة 120 دقيقة مع إضافة الكوارتز بنسبة 15% يتم الحصول على القيم المثل لمقاومة الحوامض والتي تحسنت بمقدار 86.66%، بينما تم الحصول على القيم المثل لمقاومة القواعد في العينات الخالية من إضافات الكوارتز بعد إجراء التعامل الحراري عند 600 درجة مئوية لمدة 120 دقيقة. تم أيضا استخدام موديل رياضي و تم الحصول على معادلات انحدار باستخدام برنامج ( SPSS ) والتي تكونت لغرض التنبؤ بالبيانات المختبرة لخواص الطلاء. بينت النتائج من خلال المقارنة بين القيم المتنبئة والحقيقية إن هذا البرنامج يعطي دقة تنبؤ عالية. فقد كانت دقة التنبؤ بمقدار (82.58%، و 96.7%) لمقاومة الحوامض والقواعد على التوالي.

## Introduction

**F**errous alloys are liable to corrosion, due to which their losses amount to about 10% of the world output. Different coatings are used to protect metals from corrosion. A number of coating systems are known ranging from metallic or polymeric to oxide based ceramics. Among them glass-ceramic coatings have additional advantages of chemical inertness, high temperature stability and superior mechanical properties as compared to other non-oxide coatings currently in use, for example, metallic, polymeric, etc [1]. Often, the coating systems used possess a very specific end use requirement with narrow range of selectivity. Modern engineering and technology demand such corrosion resistant coatings having a broad spectrum of effectivity under wider range of hostile environments. It is also essential that the corrosion resistant coatings should possess high resistance to thermal shock, hardness and impact damage in order to lessen the chances of

failure due to reasons other than corrosion [2]. These improved glass-ceramic coatings possessing enhanced mechanical, thermal and chemical properties may fulfill the requirements of modern engineering and technology.

Glass-ceramic deposited on a metal article and fixed in firing produces a vitreous coat which protects the article from the effect of different liquid, gaseous, and solid reactants. The constituents of the feed material depend on type of coating system (double or single coat). In double coat system there are two layers of coat: ground and cover. The ground coat is an intermediate layer between metal and cover coat and must have good adherence with substrate therefore the constituents of the charge for a ground coat include smaller amounts of oxides which promoting the adhesion with metal such as cobalt oxide (CoO), nickel oxide (NiO), and manganese oxide (MnO). The cover coat must have high chemical strength and heat resistance and good aesthetic properties therefore higher content of

silica are usually included in the composition of frit for cover coat.

The process of forming a one layer enamel (single-coat) differs from the formation of two layer enamels because the former enamel fulfills the functions of both ground and cover layers at once. Single-coat enameling allows conserving raw materials and power by reducing the coats and number of firings. The use of single-coat coating makes it possible to obtain a higher-quality surface on the articles, since the surface becomes more elastic and shock-resistant when the resulting coating thickness decreases.

This paper describes the preparation, application, and evaluation of a new corrosion resistance glass-ceramic coating which can be applied, as a single coat, directly on steel panels without prior chemical treatment. Mathematical model and investigation of the effects of quartz addition and crystallization treatment on properties of the enamel coating have been also reported in this paper.

**Zubekhin** and **Manysheva** [3] described the special features of the formation of crystalline phases in the glasses in heat treatment and the optimum regime for the formation of a glass ceramic structure. **Park and Yoon** [4] prepared glass-ceramics with well-crystallized whisker-type crystals by milling and heat treatment using fly ash from a thermal power plant and waste glass cullet. **Y. Xiong et al** [5] studied the effect of vitreous enamel coating on the oxidation behavior of Ti6Al4V and TiAl alloys at high temperature. The influence of corrosion process on spectral

properties of glassy coatings have been investigated by **Siwulski and Nocun** [6]. It was confirm that the enamel based on pigments have higher durability than enamels coloured by ion dyes.

#### **Glass and Glass-Ceramic**

Vitreous enamels are defined as glossy inorganic composition which adhere to metals by fusion and protect them against corrosive conditions [7]. Often, end use requirements have dictated the need for special coating materials with much superior properties than conventional enamels. Such coatings provide protection of the metal against high temperature and corrosive atmosphere and are commonly called glass-ceramic coatings. These are distinguished from conventional vitreous enamel coatings by the presence of suitable amounts of microcrystallites uniformly distributed in a glassy phase constituting the coating Fig.(1) [8]. The crystalline phases are produced by nucleation and crystal growth from and within the glassy phase coating material by suitable heat treatment process, so as to provide the unique combination of properties viz. high degree of resistance towards abrasion, impact, high temperature, chemical corrosion and thermal shock to prevent the failure of the substrate metals under stringent operational conditions. Glass-ceramic materials are polycrystalline solids prepared by controlled crystallization of glass. Crystallization is accomplished by subjecting the glass composition to carefully regulated heat treatment schedule which results in the nucleation and growth of crystalline phases within the glass.

The homogeneity of parent glass together with controlled conditions under which the crystals are developed, result in glass-ceramic materials having a very fine grained uniform structure free from porosity. This helps in developing high mechanical strength and good electrical insulation properties [8].

### **Experimental work**

#### **Sample preparation**

The samples used as substrate for enamel coating were rectangular billets (20mm×30mm with a thickness of 1.5mm) of low alloyed low carbon steel, their chemical composition is shown in Table (1). The sample surface is exposed to a jet of an abrasive material 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.

Before blasting, oil and drawing compounds are removed by heating the surface at 455°C to burn off the organic contaminants.

#### **Frit manufacturing**

Glass frit is the major constituent in bisque (unfired) enamel coating. Frit is the homogeneous melted mixture of inorganic materials that is used in enameling steel process. Frit is prepared by fusing a variety of minerals in a furnace and then rapidly quenched the molten material.

In this study we developed coating material which are designing for application as a single-coat on various grades of steel alloys.

The frit was prepared from reagent grade chemicals:  $\text{SiO}_2$ ,  $\text{Al}_2\text{O}_3$ ,  $\text{Na}_2\text{B}_4\text{O}_7 \cdot 10\text{H}_2\text{O}$ ,  $\text{CaCO}_3$ ,  $\text{Na}_2\text{CO}_3$ ,  $\text{ZnO}$ ,  $\text{P}_2\text{O}_5$ ,  $\text{TiO}_2$ ,  $\text{Li}_2\text{CO}_3$ ,  $\text{CoO}$ ,  $\text{NiO}$  and  $\text{CaF}_2$ . The batch was evenly blended and melted in a graphite crucible in resistance furnace at 1250 °C, the batch hold at this temperature until all the raw materials have reacted and the batch has become a homogeneous, bubble-free liquid. Then a stream of the smelted and refined molten batch is drawn from the graphite crucible and quenched into water to produce a coarse granular frit.

The coarse frit was milled by using a ball mill with alumina ball as the grinding media and screened with a mesh size of 200  $\mu\text{mesh}$ , stored in an oven at 100°C to prevent problem with moisture. The chemical composition of the frit powders is listed in Table 2.

#### **Preparation of enamel slip**

For application of the coating, the frit is further processed by mixing it with certain mill-additive to make a thick slurry called "slip" to enable its uniform and thin application on the clean metal parts by dipping technique.

The rheological properties of coating material slips are very important for ensuring proper application of the coating on the substrate. Kaolinite, borax, and water were added to form a batch of enamel slip. Refractory materials can be added to impart desired properties to the fired coating. In this work quartz was added in different amounts (5,10,15%) to study their effect on coating properties. The specific gravity of the enameling slip was measured using an electronic weighing balance and was controlled between (1.7-

1.8) by adjusting the water content. Subsequently, the slip was aged for 24 hours before enameling to improve its fluidity. Table 3 shows the weight percent composition of enamel slip.

#### Coating application

The coating material in the slip form is applied on the clean metal surface by dipping method. Dipping method is a simple and quick technique requiring no special plant, where the specimen is immersed in the enamel slip, withdrawn, allowed to drain.

After application of coating slip, the coated samples were dried in an oven at 120°C for 15min to remove moisture. The dried coated samples are then fired in a box furnace at 860°C for 10min, the coating material then fuses and reacts with the clean metal surface to form a strongly adherent coating. Subsequently, to obtaining the glass-ceramic coating, the enameled samples were held in a furnace at different temperatures ( 500, 550, 600 )°C for 60 & 120 min, respectively. The entire process of glass-ceramic coating is summarized in the flow diagram of experimental program as shown in Fig (2).

#### Inspection and testing

##### 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 diffractometer in 2θ ranging between 10° to 90° using Cu Kα radiation.

#### Acid resistance test

The acid resistance test complying with ISO: 2743. A glass-ceramic sample was exposed to solution of 11% sulfuric acid acting for 2.5 hrs at the boiling temperature[9]. The corrosion rate expressed in (mg/dcm<sup>2</sup>. day) is calculated as follows:

$$\text{corrosion rate (mdd)} = \Delta w / (A \cdot t) \dots\dots(1)$$

Where: Δw: weight loss in (mg), A: area of the specimen in (dcm<sup>2</sup>),

t: time in day.

#### Alkali resistance test

The alkali resistance of the as-fired and heat treated sample are performed In accordance with ISO: 2745, the coated samples were treated with solution of 10% NaOH for 2 hrs at the boiling temperature[10]. The corrosion rate was calculated from equation (1)

#### Mathematical modeling

A statistical model for the prediction of the acid and alkali 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 4, while the dependent variables (response functions ) were acid corrosion rate (C) and alkali corrosion rate (K). All original 28 samples within experimental data shown in Table 5 were randomly divided into two data sets; the training data set and the testing data set. The training data set contained 20 samples which were used to build a prediction model as shown in Table 6 and the testing data set contained 8 samples which were used to test the flexibility

and the validity of the prediction model as shown in Table 7.

## **Results and discussion for experimental work**

### **Thickness**

Thickness of the as-fired and crystallization treated samples is nearly unchanged. Coating thickness of the enameled specimens was in range of (0.25-0.35mm). Thin layer (<0.35mm) reduce the defects of the coating and it is suitable to comply with heat transfer requirement of waste heat recovery systems.

### **X-ray diffraction**

The coating has been characterized by X-ray diffraction. It was found that the main crystalline phases formed on the coating layers were ( $\text{Li}_2\text{SiO}_3$ ,  $\text{Li}_2\text{TiSiO}_5$ ,  $\text{NaAlSi}_2\text{O}_6$ , and  $\text{SiO}_2$ ) which are the main phases in the glass-ceramic. X-ray diffraction show that, the higher the crystallization temperature, the longer the crystalline phases peak became. The same trend has been observed with increasing the crystallization time.

The results also indicates that the quartz addition has no much effect on the formed crystalline phases, only on intensity of quartz peak. The X-ray diffraction patterns for 15%quartz addition samples treated at different heat treatments are shown in Fig 3.

### **Acid corrosion resistance**

The tests of acid resistance property of coated samples were performed in term of loss in weight value against boiling  $\text{H}_2\text{SO}_4$  solution 11% for 2.5 hrs. Fig (4) shows the relation between corrosion resistance rate of the enamel as a function of crystallization temperature in different quartz addition at different

crystallization time (a) 60min, (b) 120min. It is evident from figures that the acid resistance of the heat treated coating increases with increasing crystallization temperature and times. This is due to increasing transformation of the vitreous enamel into crystalline phases during heat treatment. This crystalline phases make a complex and strong network which acts as a physical barrier between the environment and the metal substrate.

The same trend has also been observed with quartz addition for the coating. It was found that the quartz added strongly influences the acid resistance property of the resultant coating, where the acid resistance improved with increasing the amount of mill added quartz. This can be possibly due to existence of a silicic acid network formation resulting from initial reaction of acids on excess  $\text{SiO}_2$  within the coating surface which prevent further penetrating action of acids.

### **Alkali corrosion resistance**

Figure (5) presents the results of the effects of quartz addition and heat treatment conditions on alkali corrosion rate of the resultant coating. It was found that the alkali resistance decreases with increasing quartz addition. This is explained by the fact that the kinetics of failure of the enamel in alkaline solutions have a different character in principle to that in acids. Accordingly, under the action of a hot alkaline solution the enamel surface is pitted due to the presence of acidic oxides in its composition. However, the corrosion resistance of the enamel does not depend on the amount of the acids alone, but also

on the crystallization conditions. It is evident from figure that the corrosion losses of enamel in alkali solution is improved when crystallization temperature and time are increased. This improving is associated with the presence of crystalline phases ( $\text{Li}_2\text{SiO}_3$ ,  $\text{Li}_2\text{TiSiO}_5$ ), which have an extremely corrosion resistance especially for alkali media.

### Results and discussion of mathematical model

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

$$C = 47.716 - 6.157X_1 + 0.017X_2 + 0.005X_1X_2 - 0.086X_1X_3 + 0.239X_1^2 - 4 \times 10^{-7}X_1^2X_2^2 + 1.4 \times 10^{-5}X_1^2X_3^2 - 3 \times 10^{-10}X_2^2X_3^2 - 6 \times 10^{-11}X_1^2X_2^2X_3^2 \dots\dots\dots(2)$$

$$K = 3.097 + 0.085X_1 - 0.001X_2 + 0.021X_3 + 5.6X_2X_3 + 0.001X_1X_3 - 5 \times 10^{-5}X_2X_3 - 1 \times 10^{-6}X_1X_2X_3 \dots\dots(3)$$

Where :-

C : acid corrosion rate(mdd), K : alkali corrosion rate(mdd),  $X_1$ : quartz addition(%),  $X_2$ : temperature of heat treatment( $^{\circ}\text{C}$ ),  $X_3$ : holding time(min).

The values of the multiple correlation coefficient R, that tell us how strongly the multiple independent variables are related to the dependent variable, were (0.926, & 0.981) for equations (2, & 3) respectively.

The result of average percentage deviation ( $\Phi$ ) shows that the training data set (m=20) were (17.827%, and 4.495%)

for (acid corrosion rate, and alkali corrosion rate) respectively and the testing data set (m=8) were (17.42% , 3.26875% ) for (acid corrosion rate, and alkali corrosion rate) respectively .

This means that the statistical model could predict the corrosion properties with about (82.17%, and 95.5%)accuracy of the training data set and approximately (82.58%, and 96.7%) accuracy of the testing data set for (acid corrosion rate, and alkali corrosion rate) respectively .

Figures (7, and 8) 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.

### Conclusions

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

- The coating material can be applied as a single coat by simple vitreous enameling process on selected steel alloy.
- Heat treatment increases the corrosion resistance of the resultant coatings in all cases.
- The quartz addition improves the acid corrosion resistance, but decreases the alkali corrosion resistance.
- The proposed ceramic coating material possesses reasonable low melting temperature (1200-1250 $^{\circ}\text{C}$ ) and processing 860 $^{\circ}\text{C}$

temperature and does not required prior chemical treatment of surface for application, thus leading to less cost and energy consumption during preparation and application.

- The formation of different crystalline phases in resultant coating depends on initial composition, and thermal treatment conditions.
- The multiple regression models could predict the acid and alkali resistance properties with higher accuracy for different quartz addition, and thermal treatment conditions.

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

Element	C	Mn	Ni	Al	Co	Nb	W	Fe	Si	Cr	Mo	Cu	Ti	V	Pb
Low carbon low alloyed steel (%wt)	0.18	0.12	1.35	0.05	0.05	0.05	1	94	0.05	1.3	0.4	<0.05	<0.05	<0.05	<0.05



**Table (2) Chemical composition and weight percent of frit**

Compound	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	B <sub>2</sub> O <sub>3</sub>	Na <sub>2</sub> O	P <sub>2</sub> O <sub>3</sub>	ZnO	TiO <sub>2</sub>	CoO	NiO	CaO	Li <sub>2</sub> O	F <sub>2</sub>
%(wt)	52-55	0.5-2	12-16	6-9	3-5	2-4	1-5	1-2	0.5-1	2.5-4	2-5	1-3

**Table (3) Weight percent composition of coat slip.**

Compound	Frit	Clay	Borax	Quartz	Water
Weight (wt%)	100	7	1	0,5,10,15	50

**Table (4) Definition and values of independent variables used in regression equation**

Designations of independent variable	Name of variable	Value
$X_1$	quartz addition (% wt)	0,5,10,15
$X_2$	temperature of heat treatment (°C)	0.60,120
$X_3$	holding time (min)	0,500,550,600

**Table (5):Original experimental data for predicted and measured acid corrosion rate.**

No	Quartz added (%)	Temp. (C°)	Time (min.)	Measured acid corrosion rate (mdd)	Predicted acid corrosion rate (mdd)	Measured alkali corrosion rate (mdd)	Predicted alkali corrosion rate (mdd)
1	0.00	0.00	0.00	48.00	47.71647	3.10	3.09732
2	0.00	500.00	60.00	40.25	27.71071	2.80	2.52342
3	0.00	500.00	120.00	24.20	22.33039	2.40	2.39615
4	0.00	550.00	60.00	13.45	22.91661	2.15	2.34219
5	0.00	550.00	120.00	11.80	17.37128	1.90	2.07837
6	0.00	600.00	60.00	10.70	11.72113	1.80	1.97975
7	0.00	600.00	120.00	9.60	5.79865	1.60	1.44280
8	5.00	0.00	0.00	21.45	22.91379	3.55	3.52079
9	5.00	500.00	60.00	11.30	14.69967	3.25	2.98508
10	5.00	500.00	120.00	11.30	11.32982	2.90	2.88210
11	5.00	550.00	60.00	10.75	13.14330	2.80	2.78712
12	5.00	550.00	120.00	10.45	11.58300	2.40	2.52947
13	5.00	600.00	60.00	9.95	8.20045	2.30	2.39122
14	5.00	600.00	120.00	9.35	9.96921	1.90	1.82421
15	10.00	0.00	0.00	10.45	10.07698	3.75	3.94426

16	10.00	500.00	60.00	10.25	8.70973	3.55	3.44674
17	10.00	500.00	120.00	10.05	6.70435	3.35	3.36806
18	10.00	550.00	60.00	9.75	8.83119	3.25	3.23206
19	10.00	550.00	120.00	9.45	8.90976	2.90	2.98058
20	10.00	600.00	60.00	8.85	6.57547	2.85	2.80269
21	10.00	600.00	120.00	8.10	9.80327	2.30	2.20562
22	15.00	0.00	0.00	9.70	9.20603	4.45	4.36772
23	15.00	500.00	60.00	8.95	9.74088	4.15	3.90840
24	15.00	500.00	120.00	8.55	8.45398	3.85	3.85401
25	15.00	550.00	60.00	8.20	9.98028	3.55	3.67699
26	15.00	550.00	120.00	7.55	9.35156	3.30	3.43168
27	15.00	600.00	60.00	7.10	6.84621	3.00	3.21416
28	15.00	600.00	120.00	6.40	5.30080	2.70	2.58703

Table (6): Training data for acid and alkali corrosion rate.

No.	Quartz added (%)	Temp. (C°)	Time (min.)	Measured acid corrosion rate (mdd)	Measured alkali corrosion rate (mdd)
1	0.00	0.00	0.00	48.00	3.10
2	0.00	500.00	60.00	40.25	2.80
3	0.00	550.00	60.00	13.45	2.15
4	0.00	600.00	60.00	10.70	1.80
5	0.00	600.00	120.00	9.60	1.60
6	5.00	500.00	60.00	11.30	3.25
7	5.00	550.00	60.00	10.75	2.80
8	5.00	550.00	120.00	10.45	2.40
9	5.00	600.00	60.00	9.95	2.30
10	5.00	600.00	120.00	9.35	1.90
11	10.00	0.00	0.00	10.45	3.75
12	10.00	500.00	60.00	10.25	3.55
13	10.00	500.00	120.00	10.05	3.35
14	10.00	550.00	60.00	9.75	3.25
15	10.00	550.00	120.00	9.45	2.90
16	15.00	0.00	0.00	9.70	4.45
17	15.00	500.00	120.00	8.55	3.85
18	15.00	550.00	120.00	7.55	3.30
19	15.00	600.00	60.00	7.10	3.00
20	15.00	600.00	120.00	6.40	2.70

Table (7): Testing data for acid and alkali corrosion rate.

No.	Quartz added (%)	Temp. (C°)	Time (min.)	Measured corrosion (mdd)	acid rate	Measured alkali corrosion rate (mdd)
1	0.00	500.00	120.00	24.20		2.40
2	0.00	550.00	120.00	11.80		1.90
3	5.00	0.00	0.00	21.45		3.55
4	5.00	500.00	120.00	11.30		2.90
5	10.00	600.00	60.00	8.85		2.85
6	10.00	600.00	120.00	8.10		2.30
7	15.00	500.00	60.00	8.95		4.15
8	15.00	550.00	60.00	8.20		3.55

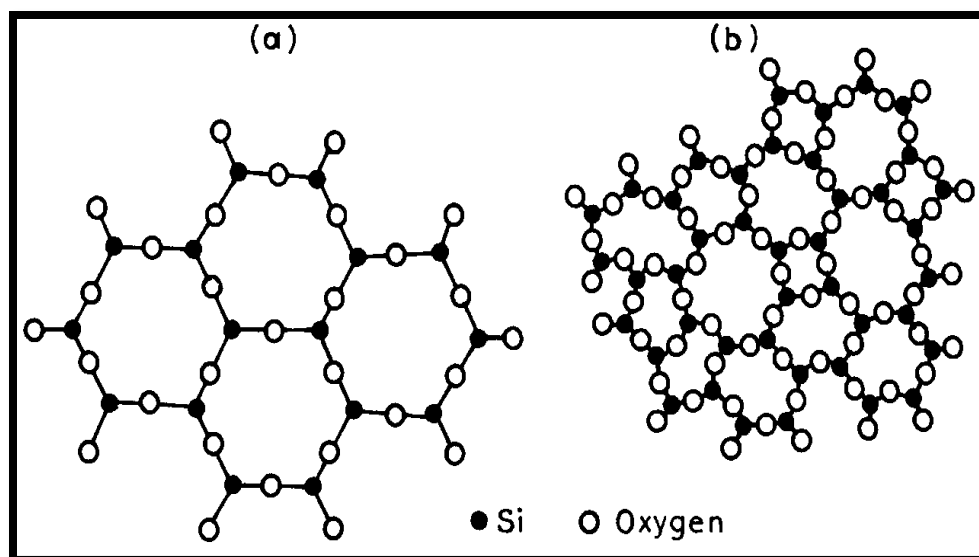


Figure (1): (a) Crystalline material (regular) and (b) glassy material (irregular)[8].

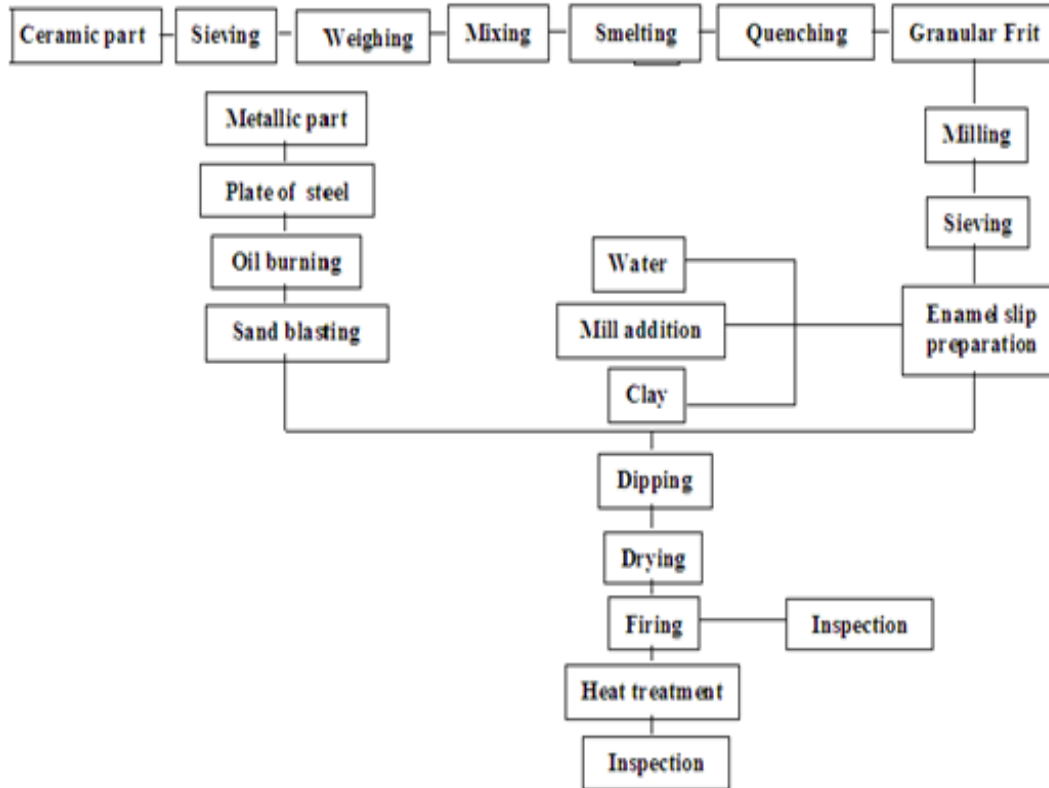


Figure (2) Flow diagram of experimental program

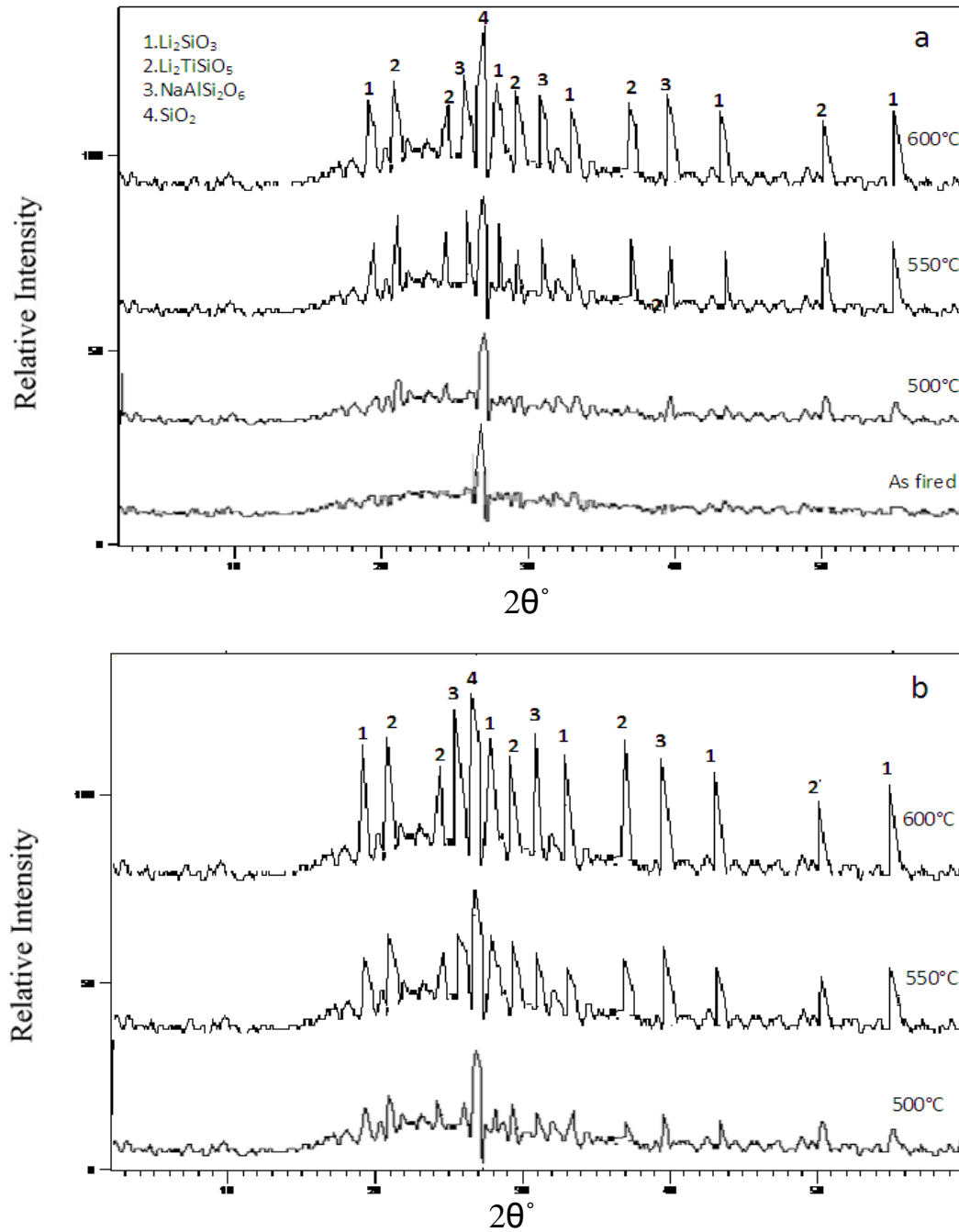


Figure (3) XRD patterns of samples (group D) for different temperatures  
at two times a: 60 min, b: 120 min

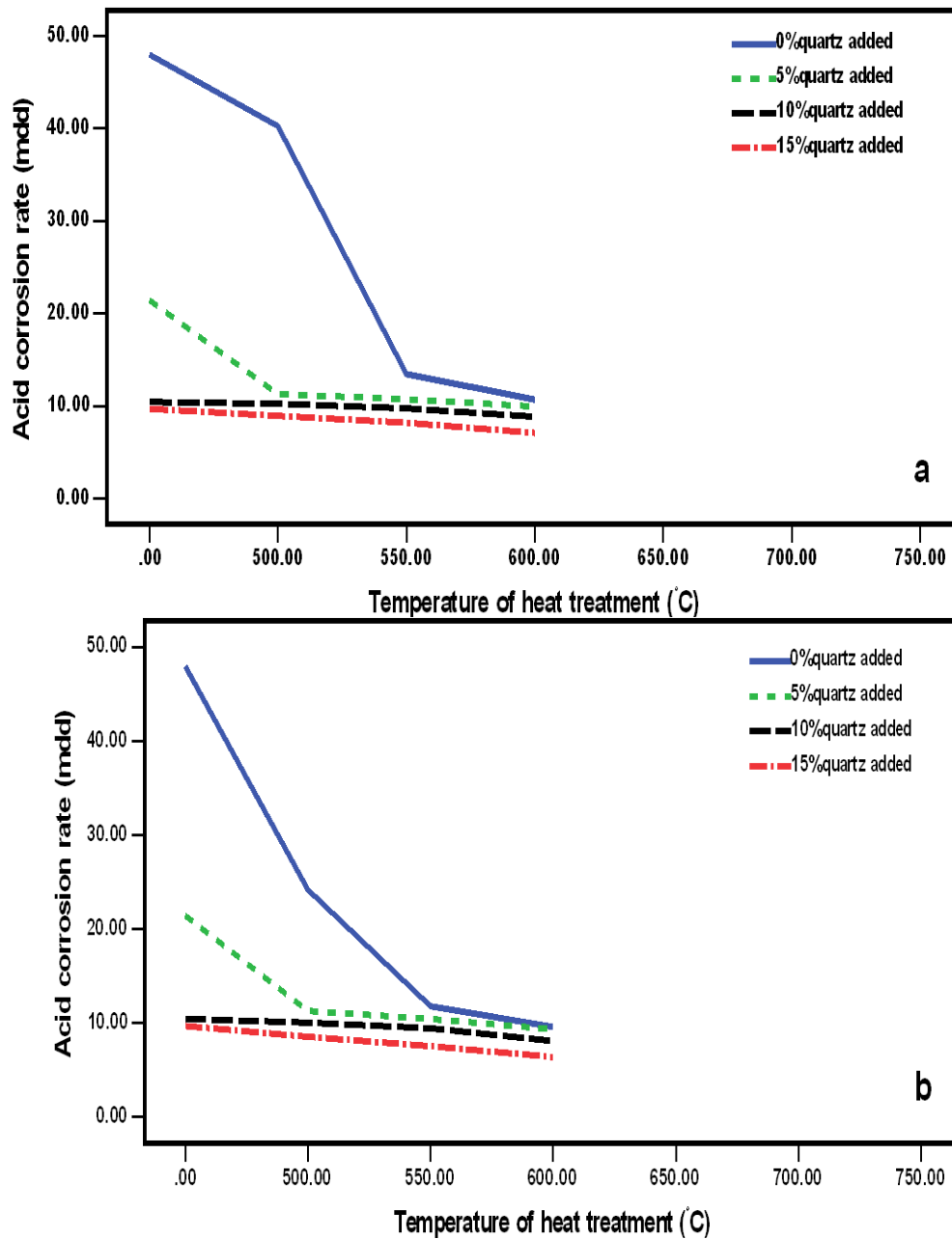


Figure (5) Relation between acid corrosion rate of the coating and crystallization temperature in difference addition of quartz at different crystallization times: (a) 60min, (b) 120min.

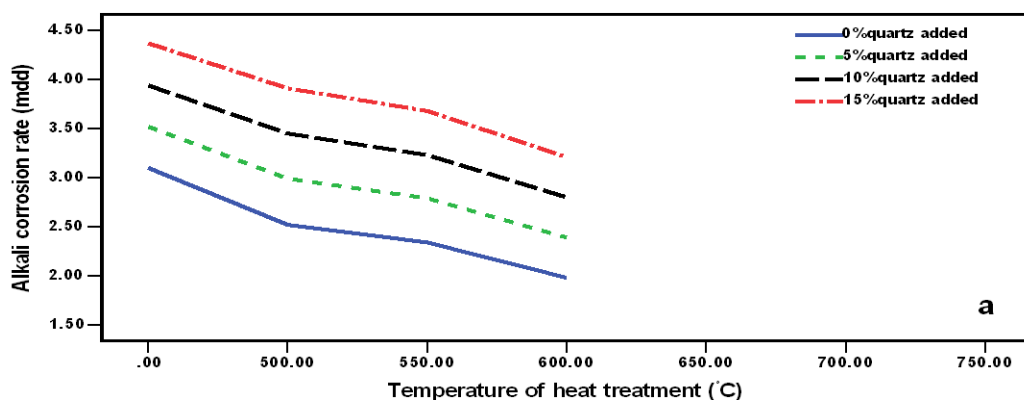


Figure (6) Relation between alkali corrosion rate of the coating and crystallization temperature in difference addition of quartz at different crystallization times: (a) 60min, (b) 120min.

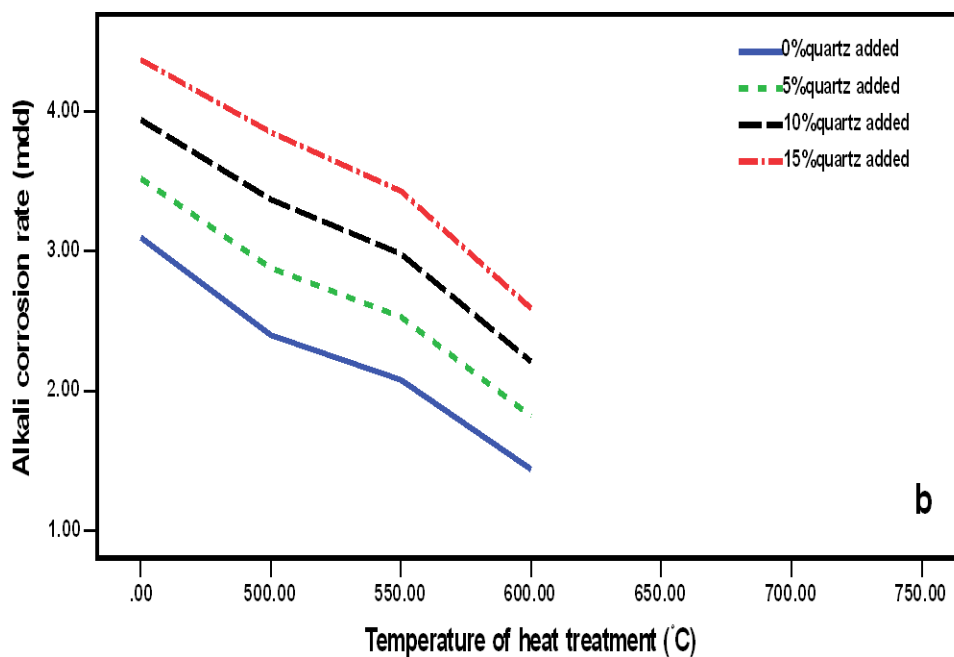
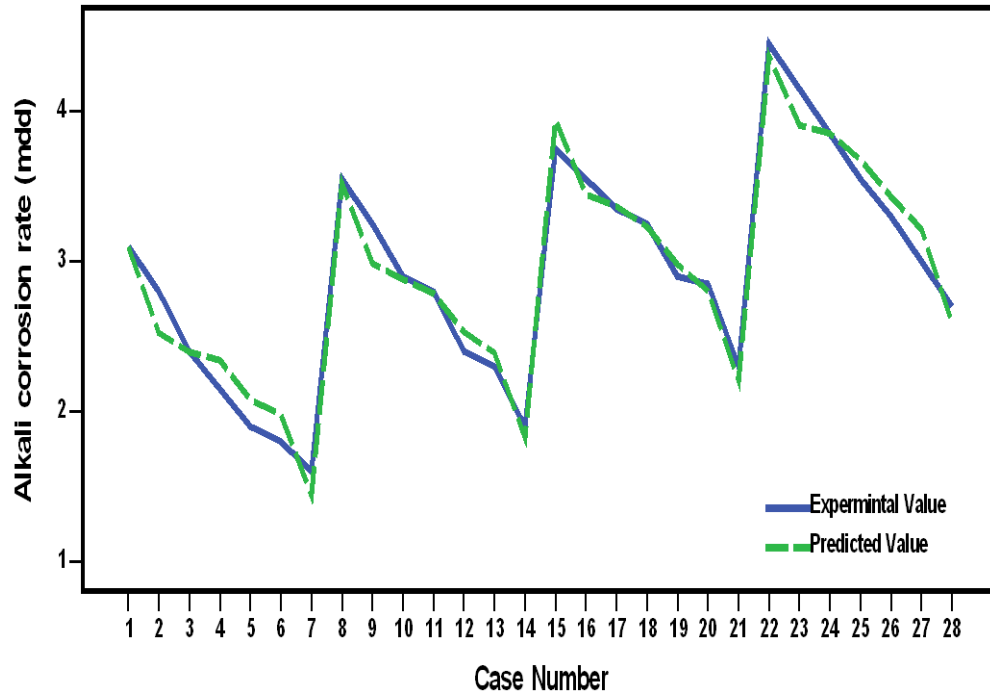


Figure (7): Comparison between measured and predicted values for the experimental data for acid corrosion rate.



**Figure (8): Comparison between Measured and Predicted values for the experimental data for alkali corrosion rate.**