

Studying and Modeling the Effects of Quartz Addition and Heat Treatment on Mechanical Properties of Glass-Ceramic Coating

Dr. Ali H. Ataiwi

Material Engineering Department, University of Technology, Baghdad
ataiwiali1@yahoo.com

Dr. Ibtihal A. Mahmood

Mechanical Eng. Dept., University of Technology, Baghdad

Jabbar H. mohammed

Material Engineering Department, University of Technology, Baghdad

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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, using the dipping technique on metal substrate. The coating are designed for application on various 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 showed the suitability of this coating for protection the metal substrate which was used in present work. The results of tests also indicated that the mechanical properties (hardness, adhesion strength, and thermal stability) of resultant coating were greatly improved by both addition of quartz into enamel frit and heat treatments for all cases, this is attributed to the formation of complex network from crystalline phases (Li_2SiO_3 , $\text{Li}_2\text{TiSiO}_5$, $\text{NaAlSi}_2\text{O}_6$, and SiO_2) which are the main phases in the resultant glass-ceramic coating. It has been found that the heat treatment at 600 °C for 120min with 15% quartz addition brought the optimum values for (hardness, adhesion strength, and thermal stability) of resultant coating which are improved by (70.58%, 33.84%, and 39.68%) respectively. Mathematical modeling is implemented and regression equations are obtained using (SPSS) software to predict the experimental data for mechanical properties of resultant coating. Comparing the predicted and measured values gives high prediction accuracy. The accuracy of prediction has been (98%, 98.5%, and 97.4%) for (hardness, adhesion strength, and thermal stability) respectively.

Keywords: enamel, glass-ceramic, SPSS software

دراسة ونمذجة تأثيرات اضافة الكوارتز والمعاملة الحرارية على الخواص الميكانيكية لطلاء الزجاج السيراميكي

الخلاصة:

في هذا العمل تم تطوير طلاء سيراميكي جديد واستخدامه، كطبقة طلاء واحدة بدون معاملة كيميائية مسبقة للسطح، باستخدام طريقة التغطية على المعدن الاساس. صمم الطلاء للاستخدام على الانواع المختلفة للفولاذ المنخفض السبك، المادة الاساس المختارة كانت فولاذ منخفض السبك منخفض الكربون (0.2%) كربون. تم استخدام معاملات حرارية مختلفة عند درجات حرارة (500، 550، 600) درجة مئوية وبازمان مختلفة (60، 120) دقيقة مع اضافات كوارتز بحدود (0-15)% للحصول على الخواص المثلى لطلاء السيراميك الزجاجي. تم توصيف هذه الطلاءات باستخدام تحليل حيود الاشعة السينية حيث اثبتت النتائج وجود عدد من الاطوار البلورية الدقيقة والتي تكونت خلال المعاملات الحرارية. بينت النتائج ملائمة هذا الطلاء لحماية المعدن الاساس المستخدم في هذا العمل. كذلك بينت نتائج الاختبارات ان الخواص الميكانيكية (الصلادة، متانة الالتصاق، والاستقرارية الحرارية) للطلاء الناتجة قد تحسنت بصورة كبيرة من خلال كلا من اضافة الكوارتز الى مادة الطلاء (الفرت) وكذلك من خلال المعاملة الحرارية وفي جميع الحالات. هذا التحسن مرتبط بتكون شبكة معقدة من الاطوار البلورية (Li_2SiO_3) وقد وجد ان المعاملة الحرارية عند (600) درجة مئوية واطراف (15)% كوارتز قد جاءت بالقيم المثلى ل (الصلادة، متانة الالتصاق، والاستقرارية الحرارية) للطلاء الناتج حيث تحسنت بنسبة (58.70، 84.33، و 68.39)% على التوالي. تم استخدام موديل رياضي والحصول على معادلات الانحدار باستخدام برنامج (SPSS) للنتائج بالبيانات العملية للخواص الميكانيكية للطلاء الناتج. اعطت المقارنة بين قيم النتائج المنتبئة والمقاسة دقة تنبأ عالية. حيث كانت دقة التنبأ (98.5، 97.4، و 98.5)% لخواص (الصلادة، متانة الالتصاق، والاستقرارية الحرارية) على التوالي.

INTRODUCTION

Modern technology uses a number of surface coating materials applied using technologies viz. PVD, CVD, plasma, etc for different industrial and engineering applications. Among various coating systems, oxide based glassy and glass-ceramic coatings provide additional advantage of chemical inertness, high temperature stability and superior mechanical properties as compared to other nonoxide coatings currently in use e.g. metals, polymers, paints, rubbers, etc (1)

Technical glass-ceramic enamels represent crystallized silicate coatings, which, after suitable thermal treatment, contain a significant concentration of finely dispersed and uniformly distributed crystalline phases. These special enamels possess much better thermo-mechanical properties relative to conventional enamel coatings. Glass-ceramic enamels are characterized by high thermal stability, working ability at considerable higher temperatures, depending on their composition and structure [2]. Glass-ceramic materials are 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. [3].

Zubekhin and Manysheva [4] described the special features of the formation of crystalline phases in the glasses in heat treatment and determined the optimum regime for the formation of a glass ceramic structure. Park and Yoon [5] 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. Xiong et al [6] 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 has been investigated by Siwulski and Nocun[7]. It was confirmed that the enamel based on pigments have higher durability than enamels coloured by ion dyes.

This paper describes the preparation, application, and evaluation of a new one coat glass-ceramic coating which can be applied 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 glass-ceramic coating have been also reported in this paper.

Experimental work

Sample preparation

The samples used as substrate for enamel coating were rectangular plates (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 abrasive material (quartz sand) 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 for 15 min. to burn off the organic contaminants.

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

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 a coating material, which, is designing for application as a single-coat on various grads of steel alloys, was developed.

The frit was prepared from reagent grade chemicals: SiO₂, Al₂O₃, Na₂B₄O₇.10H₂O, CaCO₃, Na₂CO₃, ZnO, P₂O₅, TiO₂, Li₂CO₃, CoO, NiO and CaF₂. 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 using a ball mill with alumina balls as the grinding media and screened with a mesh size of 200 mesh, stored in an oven at 100°C for 15 min. to prevent problem with moisture. The chemical composition of the frit powders is listed in Table (2).

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

compound	SiO ₂	Al ₂ O ₃	B ₂ O ₃	Na ₂ O	P ₂ O ₃	Zn O	TiO ₂	CoO	NiO	CaO	Li ₂ O	F ₂
%(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

Preparation of enamel slip

For application of the coating, the frit is further processed by mixing it with certain mill-additive to make 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, and 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 hr before enameling to improve its fluidity. Table (3) shows the weight percent composition of enamel slip.

Table (3). Weight percent composition of coat slip

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

Coating application

The coating process was carried out by dipping method where the clean metal has been dipped into the slip. 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 obtain 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 Figure (1).

Inspection and testing

Coating characterization

The thickness of the resultant coating was measured by an eddy current based thickness

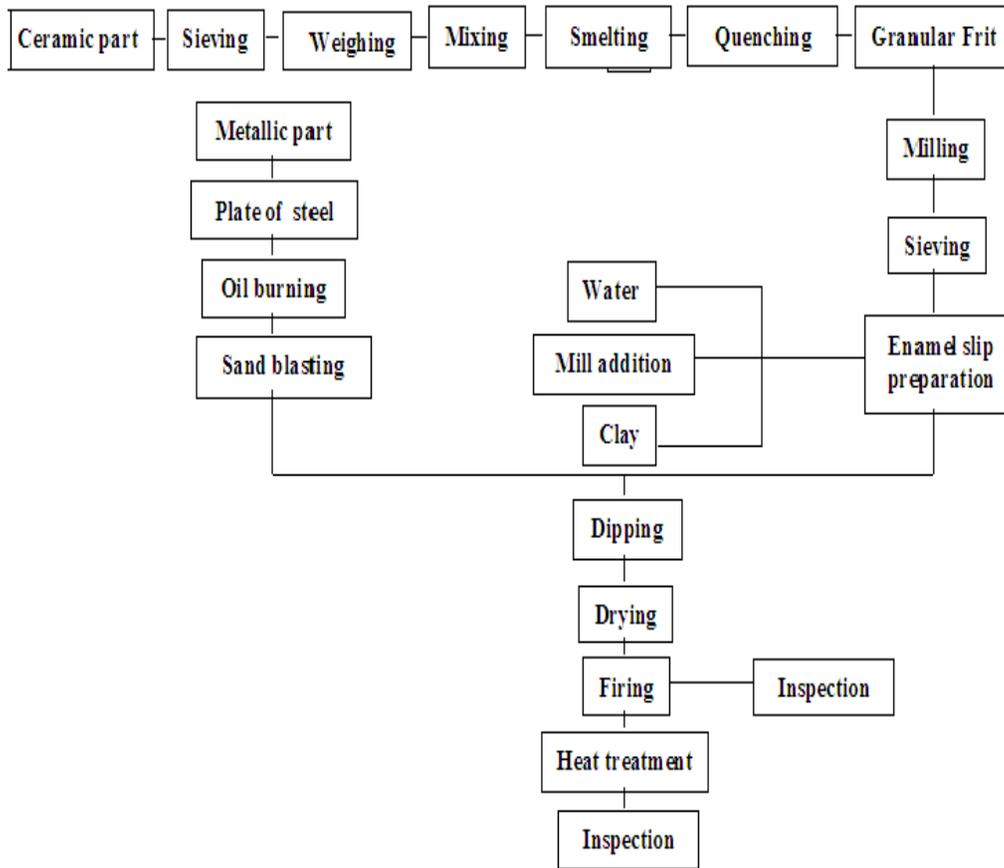


Figure 1. Flow diagram of experimental program

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) the results of this test were discussed in previous paper [8].

Microhardness measurement

The hardness of the specimens was measured with a Vickers micro-hardometer model (HVS-1000). The hardness tests were performed under an indentation load of 50 g for 20 s. In order to obtain reliable statistical data, analysis points were spaced so as to

eliminate the effect of neighboring indentations, and the hardness was evaluated by taking five indentations on each specimen and averaging of only three middle values.

Adherence strength test

In accordance with EN10209,[9] the test in Europe standard of enamel adherence strength, the enameling specimen is tested by the steel ball falling impact. Following the destruction, the adherence strength was judged according the relics of enamel on the destroyed surface. The enamel adherence strength can be graded into 1st, 2nd, 3rd, 4th and 5th grade where the 1st grade is the best. If the most of the enamel layer is removed from the steel sheet, and the surface appears silvery bright after the impact, the adherence strength is poor and 5th grade. If the most of enamel layer remains on the steel sheet, the adherence strength is excellent and 1st.

Thermal stability

A simplified method was used for these tests, whereby the enamel samples were exposed to cyclic heating up to a certain temperature, maintained the temperature for 30 min, and then quenching by pouring them into distilled water. Alternate heating and cooling was repeated several times, starting from 200 °C, whereby the samples were heated each time to a temperature 50 °C higher than the previous one.

The sample surfaces were observed and evaluated visually until the occurrence of visible surface damage. The temperature difference ΔT (°C) was determined between the maximal heating temperature, at which, after cooling with distilled water, the destruction of the coating occurred, and water temperature (20 °C).

Mathematical modeling

A statistical model for the prediction of the mechanical properties 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 the coating properties (hardness(H), adhesion strength (S), or thermal stability (ΔT)). 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).

Table (4). Definition 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 hardness, adherence, and thermal stability

No	Quartz added (%)	Temp.(C°)	Time (min.)	Measured hardness (HV)	Predicted hardness (HV)	Measured adherence (%)	Predicted adherence (%)	Measured thermal stability (C°)	Predicted thermal stability (C°)
1	0.00	0.00	0.00	492.00	492.49027	65.00	64.97079	630.00	632.96109
2	0.00	500.00	60.00	502.00	487.13511	71.00	67.41576	630.00	620.44123
3	0.00	500.00	120.00	514.00	524.57428	73.00	70.62141	680.00	685.35081
4	0.00	550.00	60.00	530.00	543.80190	70.70	69.61104	720.00	744.55834
5	0.00	550.00	120.00	620.00	598.38886	77.00	75.08805	830.00	796.14940
6	0.00	600.00	60.00	690.00	685.89967	75.00	74.00161	830.00	822.79255
7	0.00	600.00	120.00	763.00	774.78223	86.00	84.02134	830.00	847.74658
8	5.00	0.00	0.00	490.90	496.46287	67.00	66.83263	630.00	621.22512
9	5.00	500.00	60.00	508.02	495.47741	66.60	68.89552	630.00	620.86136
10	5.00	500.00	120.00	520.17	533.15321	71.00	70.78224	630.00	683.21218
11	5.00	550.00	60.00	536.36	552.55751	70.00	71.29257	720.00	742.70992
12	5.00	550.00	120.00	627.44	607.38110	72.00	75.55872	830.00	788.00218
13	5.00	600.00	60.00	698.28	695.48190	78.00	76.08666	830.00	816.40704
14	5.00	600.00	120.00	772.16	784.60108	82.00	85.11167	830.00	827.58219
15	10.00	0.00	0.00	516.60	512.09391	69.00	68.69446	680.00	688.06059
16	10.00	500.00	60.00	527.10	515.47815	68.70	70.37528	720.00	699.85292
17	10.00	500.00	120.00	539.70	553.39057	68.80	70.94307	780.00	759.64497
18	10.00	550.00	60.00	556.50	572.97156	72.00	72.97409	830.00	819.43294
19	10.00	550.00	120.00	651.00	628.03178	74.00	76.02938	830.00	858.42639
20	10.00	600.00	60.00	724.50	716.72256	79.00	78.17170	880.00	888.59296
21	10.00	600.00	120.00	801.15	806.07837	85.20	86.20200	880.00	885.98923
22	15.00	0.00	0.00	541.20	539.38337	71.00	70.55629	680.00	677.75320
23	15.00	500.00	60.00	552.20	547.13732	70.50	71.85505	680.00	701.70163
24	15.00	500.00	120.00	565.40	585.28637	76.20	71.10390	780.00	758.93490
25	15.00	550.00	60.00	583.00	605.04403	75.00	74.65562	830.00	819.01309
26	15.00	550.00	120.00	682.00	660.34088	78.00	76.50005	830.00	851.70774
27	15.00	600.00	60.00	759.00	749.62165	84.00	80.25675	880.00	883.63602
28	15.00	600.00	120.00	839.30	839.21409	87.00	87.29234	880.00	867.25342

Table (6). Training data for coating properties

No.	Quartz added (%)	Temp. (C°)	Time (min.)	Measured hardness	Measured adherence (%)	Measured acid corrosion rate (mdd)	Measured alkali corrosion rate (mdd)	Measured thermal stability (C°)
1	0.00	0.00	0.00	492.00	65.00	48.00	3.10	630.00
2	0.00	500.00	60.00	502.00	71.00	40.25	2.80	630.00
3	0.00	550.00	60.00	530.00	70.70	13.45	2.15	720.00
4	0.00	600.00	60.00	690.00	75.00	10.70	1.80	830.00
5	0.00	600.00	120.00	763.00	86.00	9.60	1.60	830.00
6	5.00	500.00	60.00	508.02	66.60	11.30	3.25	630.00
7	5.00	550.00	60.00	536.36	70.00	10.75	2.80	720.00
8	5.00	550.00	120.00	627.44	72.00	10.45	2.40	830.00
9	5.00	600.00	60.00	698.28	78.00	9.95	2.30	830.00
10	5.00	600.00	120.00	772.16	82.00	9.35	1.90	830.00
11	10.00	0.00	0.00	516.60	69.00	10.45	3.75	680.00
12	10.00	500.00	60.00	527.10	68.70	10.25	3.55	720.00
13	10.00	500.00	120.00	539.70	68.80	10.05	3.35	780.00
14	10.00	550.00	60.00	556.50	72.00	9.75	3.25	830.00
15	10.00	550.00	120.00	651.00	74.00	9.45	2.90	830.00
16	15.00	0.00	0.00	541.20	71.00	9.70	4.45	680.00
17	15.00	500.00	120.00	565.40	76.20	8.55	3.85	780.00
18	15.00	550.00	120.00	682.00	78.00	7.55	3.30	830.00
19	15.00	600.00	60.00	759.00	84.00	7.10	3.00	880.00
20	15.00	600.00	120.00	839.30	87.00	6.40	2.70	880.00

Table (7). Testing data for coating properties

No.	Quartz added (%)	Temp. (C°)	Time (min.)	Measured hardness	Measured adherence (%)	Measured acid corrosion rate (mdd)	Measured alkali corrosion rate (mdd)	Measured thermal stability (C°)
1	0.00	500.00	120.00	514.00	73.00	24.20	2.40	680.00
2	0.00	550.00	120.00	620.00	77.00	11.80	1.90	830.00
3	5.00	0.00	0.00	490.90	67.00	21.45	3.55	630.00
4	5.00	500.00	120.00	520.17	71.00	11.30	2.90	630.00
5	10.00	600.00	60.00	724.50	79.00	8.85	2.85	880.00
6	10.00	600.00	120.00	801.15	85.20	8.10	2.30	880.00
7	15.00	500.00	60.00	552.20	70.50	8.95	4.15	680.00
8	15.00	550.00	60.00	583.00	75.00	8.20	3.55	830.00

Results and discussion for experimental work
Hardness

Figure (2) shows the effects of heat treatment temperatures on the micro-hardness values with different amount of quartz addition for two crystallization time (a) 60min, (b) 120min. It is clear from the figures that the quartz addition increases the hardness value. This improving in hardness of the coating is due to incorporating a greater percentage of a hard component (quartz) into enamel. It is also obvious from figures that the hardness value of the as-fired enamel (492HV_{0.05}) is enhanced greatly by the crystallization treatment to reaches (839HV_{0.05}) at 600°C for 120 min. When the crystallization time is kept constant, hardness of the enamel coating is improved in all crystallization temperature, and at each of crystallization temperatures, the hardness of the coating increases with increasing time, as shown in Fig (3a & b), which lies in the phase transformation that occurred in coating during crystallization treatment. The hard, unique and complex microstructure of the formed crystalline phases especially the phase (NaAlSi₂O₆) [8] is responsible for the excellent micro-hardness property of the resultant heat treated coatings.

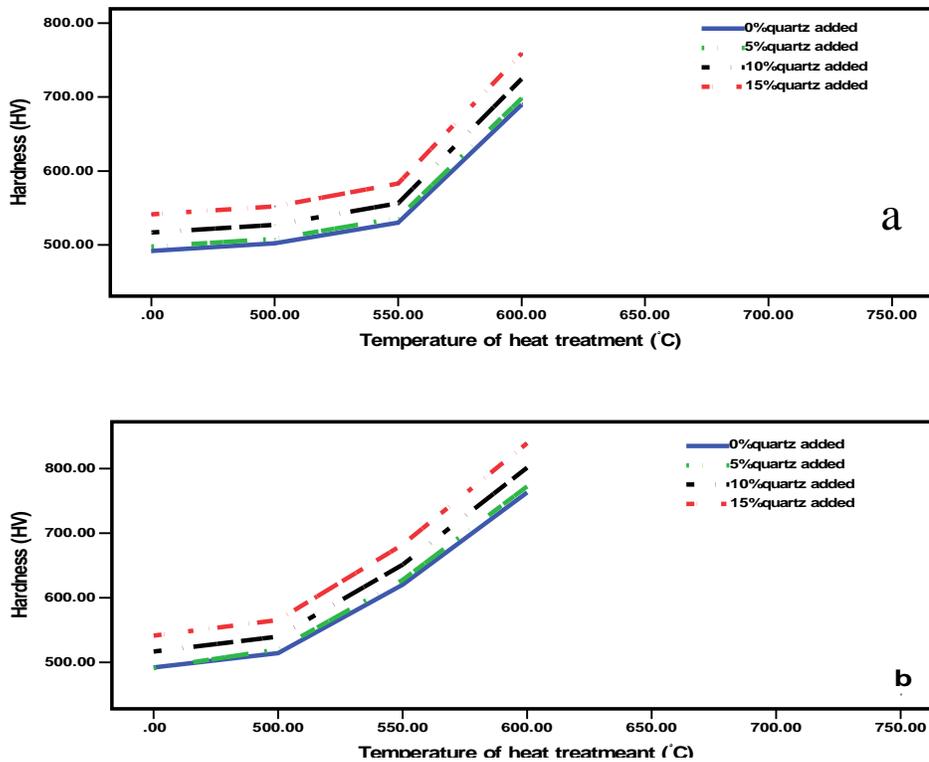


Figure (2). Relation between hardness values of the coating and crystallization temperature in difference addition of quartz at different crystallization times: (a) 60min, (b) 120min

Adhesion strength

The adhesion mechanisms are intimately related to the microstructure and morphology of the intermediate boundary between enamel layer and steel substrate. Considering that the heat treatment and addition of quartz may exert great influence on the microstructure of intermediate bonding layer, and as result on adhesion strength. Fig(3) shows the effects of heat treatment and mill addition on adherence of glass-ceramic coating to steel. It can be seen that the quartz additive improving the adhesion strength. This is can be due to the rugged interface and increasing the contact area and junction point to substrate, which are resulted by chemical corrosion during the firing stage.

At the same time, the adhesion strength keeps increasing with the rise of both crystallization temperature and time. Improving the adhesion strength by crystallization treatment can likely be explained by increasing the roughening of interface. This criterion is in accordance with mechanical theory where good adherence was associated with high density of anchor points at enamel/steel interface.

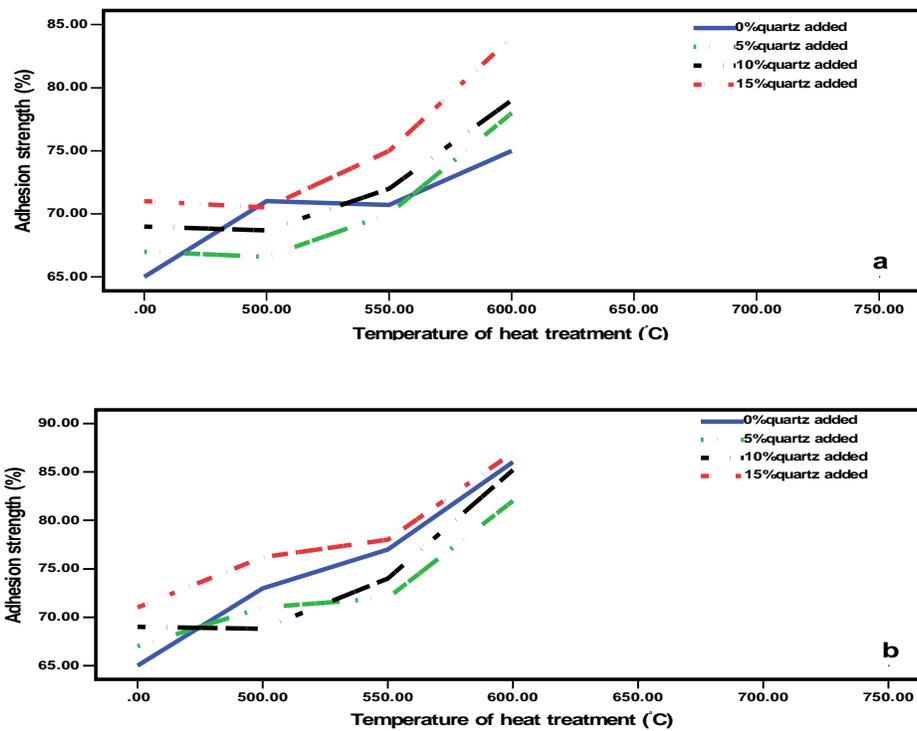
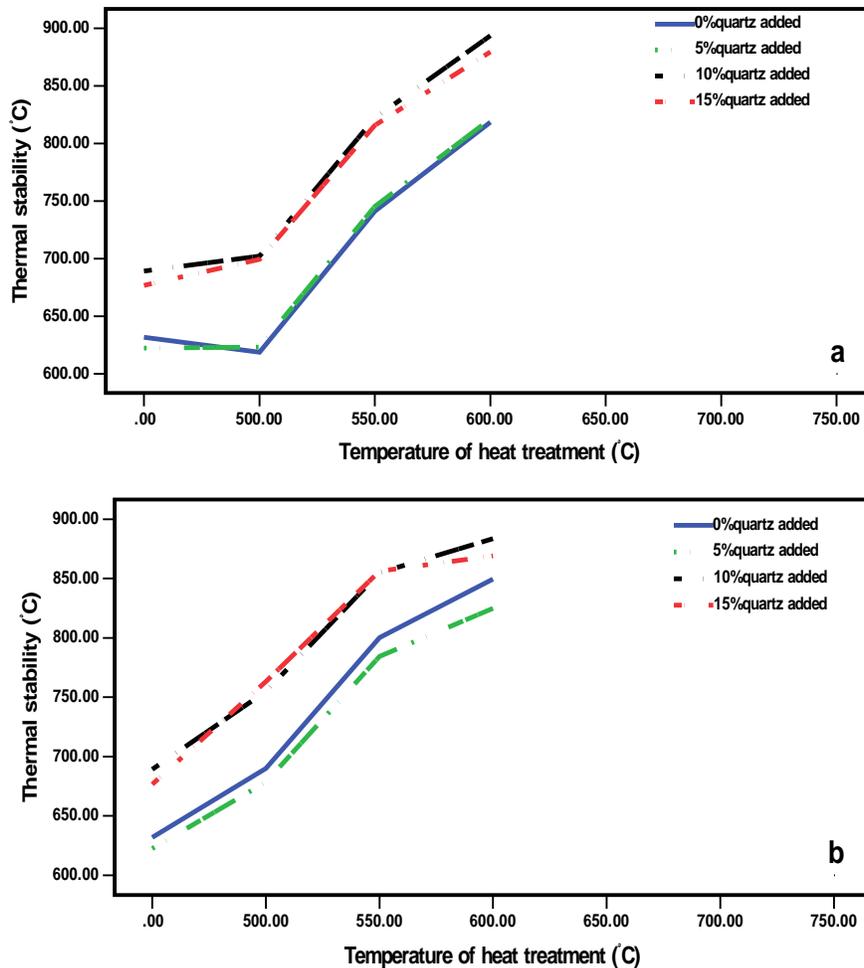


Figure (3). Relation between adhesion strength of the coating and crystallization temperature in difference addition of quartz at different crystallization times: (a) 60min, (b) 120min

Thermal stability

The results indicate the crystallization treatment has larger effect on thermal stability than quartz addition as shown in Fig(4). The obtained results show that an increase in thermal stability is realized in all the crystallization treatment, and there is no difference in results when amount of mill added quartz in range (0-5)%.

The coating which treated at 550°C for (60min) and at 600°C for (60 &120min) has maximum values of thermal stability (resist up to $\Delta T=880^{\circ}C$ without any damage). Moreover change in working temperature by 50°C doesn't effectively change the thermal stability of the coating. Only complete crystallization along with change of colour and smoothing of the coating is observed.



Figuer (4). Relation between thermal stability of the coating and crystallization temperature in difference addition of quartz at different crystallization times: (a) 60min, (b) 120min

Results and discussion of mathematical model

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

$$H = 492.49 - 0.371X_1 - 1.223X_2 + 0.002X_1X_2 + 0.001X_1X_3 + 0.006X_2X_3 + 0.233X_1^2 - 0.002X_2^2 - 0.012X_3^2 \dots\dots\dots 1$$

$$S = 64.971 + 0.371X_1 - 0.002X_2 - 0.325X_3 - 0.008X_1X_3 + 0.001X_2X_3 + 7.2 * 10^{-6}X_1X_2X_3 \dots\dots\dots 2$$

$$\Delta T = 632.961 - 20.585X_1 + 8.265X_2 - 62.358X_3 + 0.006X_1X_2 + 0.116X_1X_3 - 0.004X_2X_3 + 4.686X_1^2 - 0.208X_1^3 - 7 * 10^{-6}X_2^3 + 0.003X_3^3 \dots\dots\dots 3$$

Where:-

X₁: quartz addition (%wt), X₂: temperature of heat treatment (°C), X₃: holding time (min), H: hardness, S: adhesion strength, ΔT: thermal stability.

The values of the multiple correlation coefficient R, that tell us how strongly the multiple independent variables are related to the dependent variable, were (0.992, 0.932, 0.974) for equations (1, 2, & 3) respectively.

The result of average percentage deviation (Φ) shows that the training data set (m=20) were (1.9485%, 2.8645%, and 1.94%) for (hardness, adhesion strength, and thermal stability) respectively and the testing data set (m=8) were (1.9325%, 1.455%, and 2.61%) for (hardness, adhesion strength, and thermal stability) respectively.

This means that the statistical model could predict the coating properties with about (98% , 97.1% , and 98%) accuracy of the training data set and approximately (98% , 98.5% , and 97.4%) accuracy of the testing data set for properties (hardness, adhesion strength, thermal stability) respectively .

Figures (5, 6, and 7) show the comparison between the predicted values and measured values of 28 original data for mechanical properties respectively 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.

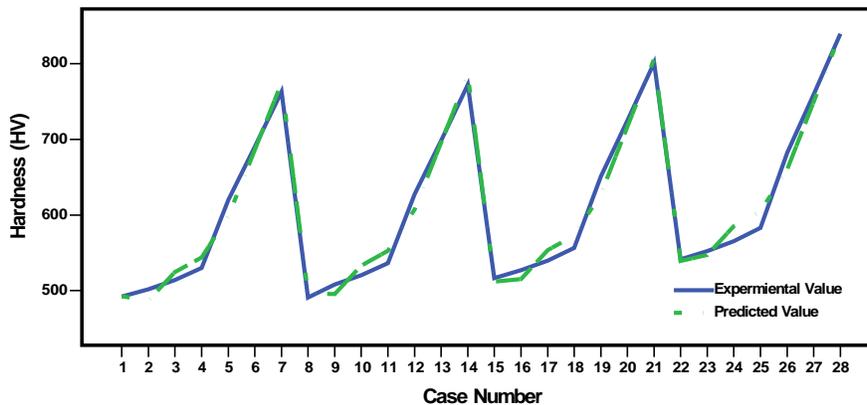


Figure (5). Comparison between Measured and the Predicted values for the experimental data for hardness

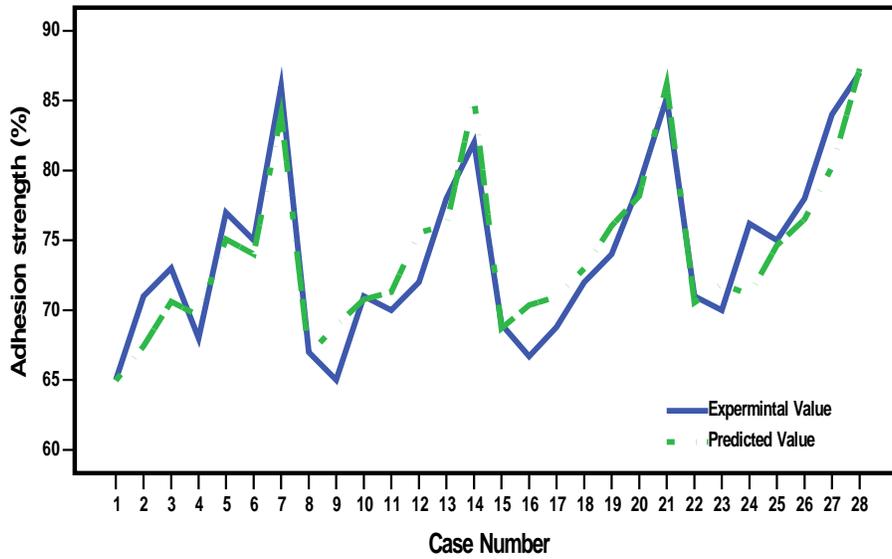


Figure (6). Comparison between Measured and the Predicted values for the experimental data for adhesion strength

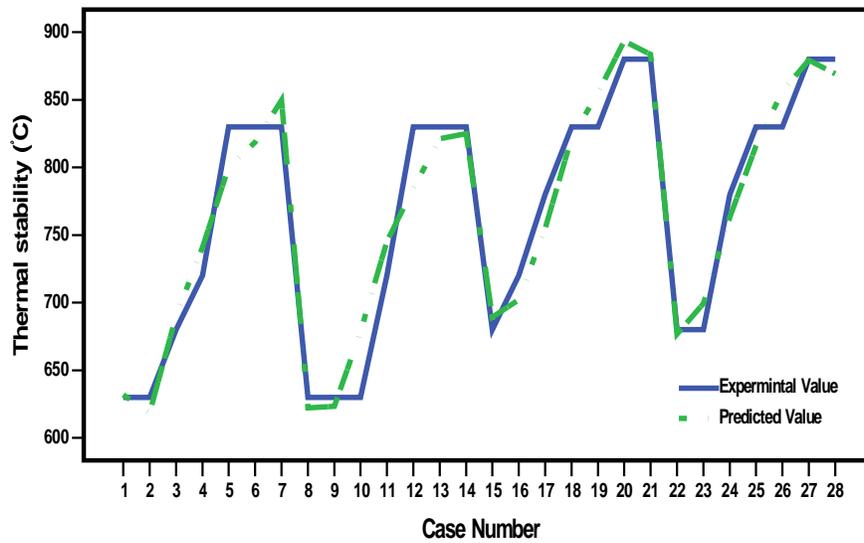


Figure (7). Comparison between Measured and Predicted values for the experimental data for thermal stability

CONCLUSION

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 and quartz addition increases the mechanical properties of the resultant coatings in all cases.
- The proposed ceramic coating material possesses reasonable low melting temperature (1200-1250°C) and processing 860°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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