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Improvement the Chemical Resistance of Furnaces Bricks for Petroleum Refineries by ZrO_2-Nano- Glass-Ceramic Coated

Abstract- *Partial Stabilized Zirconia (PSZ) was prepared from adding 3 wt % of MgO or adding 8 wt% of Y_2O_3 to 90 wt % ZrO_2 Powder and mixed by wet method, then dried and firing mixture to 1500 ℃ to obtain PSZ ceramic powder. Glass-Ceramic (Li_2SiO_3) and ($LiAlO_2$) prepared by dissolve lithium carbonate and lithium hydroxide with Nano-Silica (SiO_2) and Nano-Alumina (Al_2O_3) respectively. Those glass-ceramic mixed with PSZ in different percentage (2.5,5,7.5,10) and sprayed on furnaces bricks for petroleum refineries. An increase in the chemical resistance of the acid on the surface of the Refractory bricks was observed when coating with the glass-ceramic mixture, as well as increasing the hardness and thermal shock resistance. Lithium silicate coated specimens are more spared and homogeneous on the surface compared to lithium laminate coated.*

Keywords- *Glass-Ceramic; Coating; Partial Stabilized Zirconia; Mohs Scale; Chemical Resistance.*

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1. Introduction

Glass- ceramics are characterize as polycrystalline materials manufactured through controlled crystallization of base glass[1]. Generally, the glass-ceramic can be created by dissolving glass and changing over the substance into a uniform nucleation and growth of fine grained ceramic by controlled crystallization process in which the crystalline phases are nucleated and grown in glass by heat treatment [2].

PZT (Partially stabilized zirconia) is a mixture of zirconium oxide polymorphs, as a result of deficient cubic phase stabilized forming oxide has been added, and a cubic with metastable tetragonal ZrO_2 mixture is obtained. A smaller addition of oxide stabilizer to the zirconia will getting its structure into a tetragonal phase at sintering temperature higher than 1000 ºC , and a mixture of cubic phase and monoclinic or tetragonal phase at a lower temperature. Therefore, the PZT is also known in another way as tetragonal zirconia polycrystalline (TZP) [3]. Commonly, such PSZ contain of large than 8 mol% of magnesium oxide (MgO), 8mol% of CaO, or 3-4 Mol% of Y_2O_3. Partially stabilized zirconia is a conversion toughened material micro-crack and induced stress may be two interpretations for the toughening in PZT[4], lithium meta silicate (Li_2SiO_3) (melting point :1204 ºC)[5] as an important inorganic compound having been widely used in the field of research in depth . As the coating base material, due to good heat, burn, radiation – resistance, abrasion resistance ,moisture resistance, water resistance, weather resistance, light resistance, stain resistance and environment friendliness, lithium silicate coating can be used for inorganic building layer of material, such as maritime engineering, oil pipelines ships, bridges and architectal coating. Petroleum refinery furnaces are usually made of denes refractory concrete of fire brick in order to resist foot traffic and mechanical impact during turnarounds[6]. Stacks and breechings for most types of refinery units have similar service requirements: strength at high temperature and resistance to corrosion erosion ,and pitting. Temperature range from 250 to 815 ºC , and the flue gas may contain catalyst sulfur oxides , hydrogen sulfide , or carbon monoxide all of these emitted gases may cause a weak structure of the insulating material, whether it is a brick or cast able. this causes weakness of the mechanical properties and thus may causes the collapse of design [7]. Therefore, in this research will depend coating those surfaces with a layer of glass–ceramic to increases the chemical resistance of furnace lining.

2. Materials and Methods

I. Materials Preparation

Laboratory available ceramic materials in the micro size from: Lithium Carbonate (Li_2CO_3), Zirconia (ZrO_2) and Yttrium (Y_2O_3). The Alumina (Al_2O_3), Silica (SiO_2) and magnesia (MgO) in Nano size. Table 1 show the specification of raw materials – Li_2SiO_3 was prepared by solid state interaction method, Li_2CO_3 and SiO_2 were mixed

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by using agate mortar for 2 hours including wet mixing in acetone(CH_3COCH_3) for 1 hours.

Table 1: Specifications of materials.

Material	Source	Purity %	Particle Size
ZrO_2	Riedel de Haen	99	< 5 µm
Y_2O_3	Fixanal	99.95	< 5 µm
MgO	Nanjing Nano Tech.	99.9	30-40 nm
Li_2CO_3	BDH AnalaR	99.5	----------
SiO_2	Nanjing Nano Tech.	99.5	< 40 nm
α-Al_2O_3	Hongwu I. Group	99.9	< 80 nm
LiOH	BDH Analar	99.5	----------

The mixture was firing at 900 °C for 4h. The chemical formula for this reaction is:

$$Li_2CO_3 + SiO_2 \xrightarrow{900C^o} Li_2SiO_3 + CO_2$$

$LiAlO_2$ prepared by mixing LiOH with distilled water and Al_2O_3 at a temperature of 90 ºC. The production was realized via calcination at 800 ºC the chemical formula of this reaction is:

$$2LiOH.H_2O + Al_2O_3 \longrightarrow 2LiAlO_2 + 3H_2O$$

PSZ prepared by adding 3 wt% of MgO or 8 wt% of Y_2O_3 to ZrO_2 and mixed 4hr with distilled water after drying in oven at 90 ºC the mixture was fired at 1500 ºC at 4h. The grinding process was done on the mixture to obtain a granular size of less than 25µm. Different percentages of Li_2SiO_3 or $LiAlO_2$ were added to (Mg or Y)-PSZ as shown in table (2).

Sodium silicate (1.25 g/cm³) was added by 10 wt%, after which the surface of the medium alumina brick (using in the oil refinery furnaces) was coated using the brush. The coated sample were firing at 1200 ºC to increase the adhesion of the glass-ceramic compositions with refractory brick substrate.

II. Chemical Resistant

The chemical and physical properties of refractory brick vary according to the source. This is mainly because their composition is determined by the raw material source. Refractory bricks are considered to be of three types depend absorption and acid resistance, according to ASTM C279.

Table 2: The Specimens Composition.

Sample NO.	Y-PSZ wt%	Mg-PSZ wt%	$LiAlO_2$ wt%	Li_2SiO_3 wt%
1	97.5	0	2.5	0
2	95	0	5	0
3	92.5	0	7.5	0
4	90	0	10	0
5	0	97.5	0	2.5
6	0	95	0	5
7	0	92.5	0	7.5
8	0	90	0	10

III. Hardness (Mohs Scale)

Mohs Hardness test is one of the most important test for identifying mineral specimens. Moh's hardness scale include the test compares the resistance of a specimens to being scratched by ten reference minerals. This test was done using Rocks minerals samples specimens geology collection (24) and hardness (Mohs) kit from USA.

IV. Thermal Shock Resistance

Low thermal shock resistance is weakest points of brittle ceramic materials. Thermal shock resistance depended fracture toughness, young modulus, poisson’s ratio, thermal conductivity, thermal expansion coefficient. Stresses can be found due to the temperature difference between surface and the center of a specimen after quenching by cooled with water or heated rapidly [8]. Thermal shock test is aimed to find the site in which the specimen broken. The thermal shock test was used according to ASTM C 385-58 standard. During the experiment, samples were heated at particular temperature and waiting to regular distribution of temperature and put directly into cold water to equip thermal shock. Thermal cycling was repeated by heating 20 ° C in each step up to 300 ° C, and each step has been in control specimens until up to 1000 ° C [9].

V. Brick Substrate

The substrate that was coated with glass-ceramic layer is the medium alumina bricks (62 % Al_2O_3), supplied to Iraqi Ministry of Oil (North Oil Company), and used in lining the burner of the furnaces of the refining Units.

3. Results and Discussion

The morphology structure of specimens coated with glass-ceramic is illustrated in Figure 1, it is noticed that the distribution of glass on the surface was more uniform in the case of (Mg-PSZ, Li_2SiO_3) coating compared with the coating of (YPSZ, $LiAlO_2$) due to high melting temperature of (Y-PSZ, $LiAlO_2$). The coating layers showed different resistance against the strong acid (Sulfuric acid H_2SO_4: sp gr 1.706, or 78 weight % 60° Baumé) . The (Y-PSZ, $LiAlO_2$) specimens showed that the chemical resistance was better than the (Mg-PSZ, Li_2SiO_3). Figure 2 shows the change in color of coating layer appeared for the specimens coated (Mg-PSZ, Li_2SiO_3), and it is possible that the reason is the added MgO to partially stabilized zirconia (PSZ), where, Chemically, the MgO is classified as basic refractories. Generally, No significant deformities or micro cracks in the structure of the coatings were observed. Enhance the chemical resistance of the coating layer due to the use of ceramic materials characterized by high chemical resistance such as ZrO_2, Al_2O_3 and SiO_2.

Figure 3 show increasing the hardness (Mohs scale) with increasing the percentage of glass-ceramic additives. Mohs Scale values ranged from 6.5 to 8.5 when adding (Li_2SiO_3) glass-ceramic materials. While the Moh's scale values when adding ($LiAlO_2$) between 5 to 7.5. High melting temperature of ($LiAlO_2$)(1700 ^{o}C) comparing with (Li_2SiO_3) was the reason for decrease hardness, due to glass phase decreasing.

Mohs scale values increases means more resistant of material to scratch, and this leads to decrease in the coefficient of friction and thus increase wear resistance. Increasing scratch resistance of coated surfaces means increased the resistant of refractory bricks to damage and reduce the pitting on refractory surfaces, because the surfaces of these refractory materials exposed to the collision of gas molecules and other volatile materials from the refining processes.

Thermal expansion coefficient is one of the most important factors affecting the success of glass-ceramic coatings, because the compatibility between the thermal expansion coefficient of the coating layer and the base material is also necessary to increase the thermal shock resistant of coating layer. From different references it was found that the values of thermal expansion coefficient for glass-ceramic composition are (10-11×10^{-6} K^{-1} for $Li_2O–ZrO_2–SiO_2–Al_2O_3$ and alumina bricks) [10], and 6.5×10^{-6} K^{-1} for $LiAlO_2$ [11]. This convergence in the values of thermal expansion coefficient is the main reason behind the survival of glass-ceramic structure without cracking after exposure to thermal shock at 1000 ^{o}C. Figure 4 shows a surface morphology of the coating layer after thermal shock experiment.

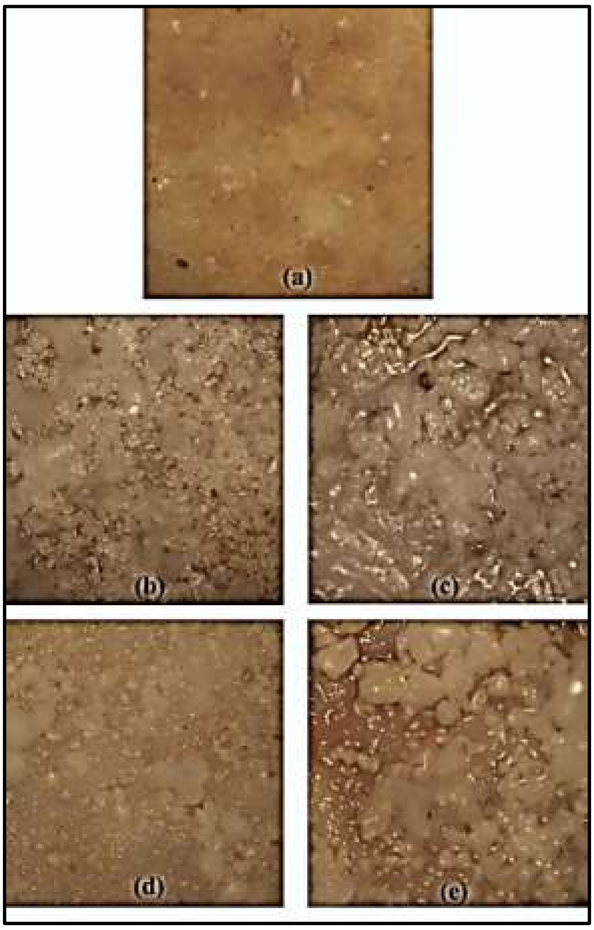

Figure 1: The Glass-Ceramic coating layers: (a) The surface of brick without coating, (b) (Mg-PSZ, 2.5 wt% Li_2SiO_3), (c) (Mg-PSZ, 10 wt% Li_2SiO_3), (d) (Y-PSZ, 2.5 wt% $LiAlO_2$), (e) (Y-PSZ, 10 wt% $LiAlO_2$).

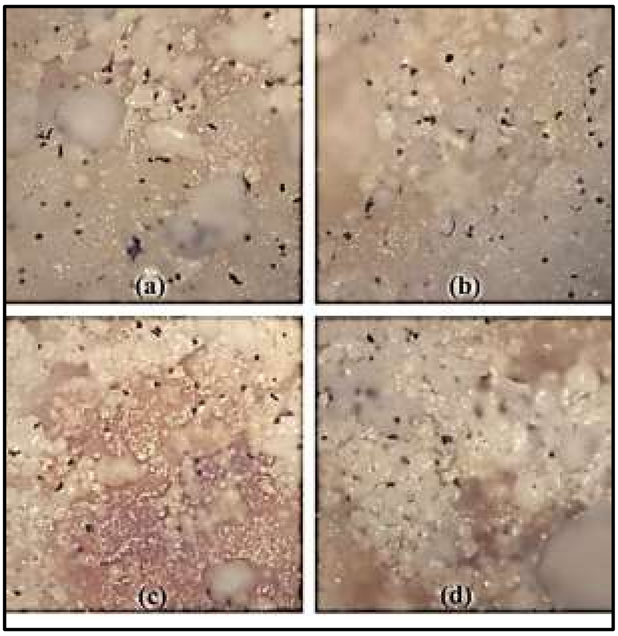

Figure. 2: The Glass-Ceramic coating layers after chemical attack: (a) (Y-PSZ, 2.5 wt% $LiAlO_2$), (b) (Y-PSZ, 10 wt% $LiAlO_2$) (c) (Mg-PSZ, 2.5 wt% Li_2SiO_3), (d) (Mg-PSZ, 10 wt% Li_2SiO_3).

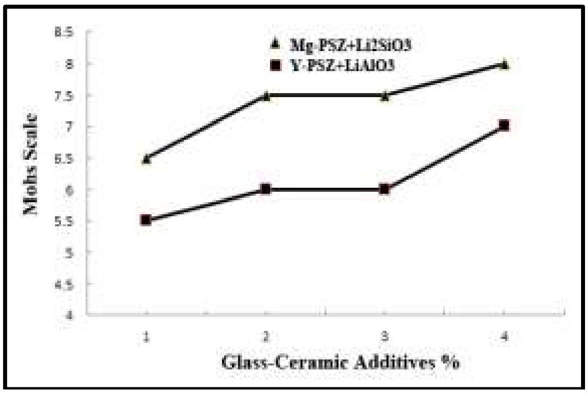

Figure 3 :Hardness (Mohs Scale) of Glass-Ceramic coated with various Li_2SiO_3 and $LiAlO_2$ additives.

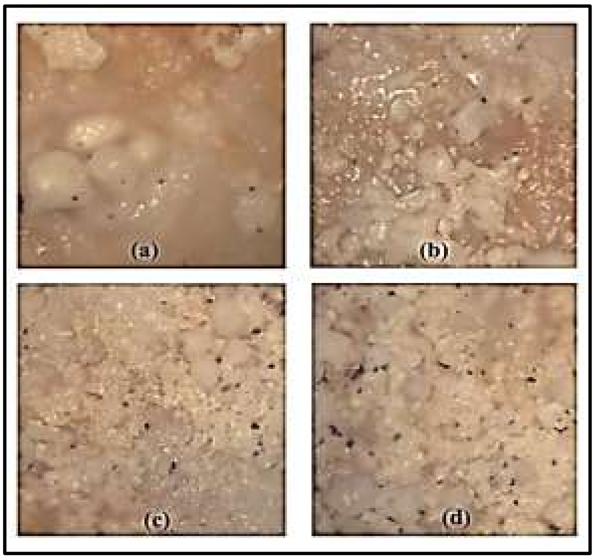

Figure 4: The Glass-Ceramic coating layers after Thermal Shock : (a) (Y-PSZ, 2.5 wt% $LiAlO_2$), (b) (Y-PSZ, 10 wt% $LiAlO_2$) (c) (Mg-PSZ, 2.5 wt% Li_2SiO_3), (d) (Mg-PSZ, 10 wt% Li_2SiO_3).

4. Conclusions

Excellent chemical resistance layer coating, make it possible to use this type of glass-ceramic coatings in the treatment and improve the surfaces of refractories and used in the construction of oil refining furnaces to be more resistant to acidic vapors emitted from oil refining processes.

Increasing scratch resistance when adding glass ceramic meaning improve the resistance of refractories to pitting from the collision of gases and volatile materials emitted during furnaces operation. Convergence in the thermal expansion coefficients of the components of the coated materials and the substrate was the cause for resisting glass- ceramic coating to thermal shock.

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