The influence of magnesia addition and sintering temperature on the properties of synthesized electrical porcelain

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ABSTRACT

The influence of magnesia (MgO) content at different sintering temperatures on the physical and dielectric properties of synthesized electrical porcelain was investigated. Feldspar is one of the major components of porcelain, since feldspar is not available in Iraq, so it synthesized from Iraqi raw materials. MgO was progressively added in the range (2-30 wt. %) into electrical porcelain. The composed bodies were sintered at temperature in the range (1200-1350 °C). Physical properties such as bulk density, open porosity beside the Vickers microhardness and dielectric properties such as dielectric constant, loss tangent and dielectric loss factor were measured. The improvement of these properties could associate with an increasing of MgO additive and sintering temperatures. Bulk density, microhardness and dielectric constant are increased with the increasing of MgO content and sintering temperatures. The results reveal that the highest value of density at 30 wt. % MgO and at 1350 °C for density, hardness and dielectric constant, while it has the lowest value for porosity and loss tangent.

Keywords: Porcelain, Feldspar, MgO, Dielectric constant, Microhardness; loss tangent.

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تاثير إضافة المغنيسيا ودرجة حرارة التلبيد على خواص البورسلين الكهربائي المحضر.

تم دراسة تأثير محتوى المغنيسيا ودرجة حرارة التلبيد على الخواص الفيزيائية والعزلية للبورسلين المحضر. يعتبر الفلدسبار أحد المكونات الرئيسية للبورسلين حيث أن هذه المادة غير متوفرة في العراق كمادة أولية. لذا نطلب تصنيعها من مواد محلية لغرض استخدامها في تصنيع البورسلين. تم إضافة نسب من المغنيسيا ضمن المدى (2-30) %

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INTRODUCTION:

Electrical porcelain has long been used as an electrical insulator in both transmission and distribution lines of electricity. The main properties exhibited by this material are: high mechanical strength, low dielectric loss, superior electrical properties, good creep resistance at room temperature and high corrosion resistance [1]. Triaxial porcelains are primarily composed of clay, feldspar and filler materials, usually quartz or alumina. The clay gives plasticity to the ceramic mixture, quartz maintains the shape of the formed article during firing and feldspar serves as flux.

The three constituents place electrical porcelain in the phase system \([(k, Na)_2 O – Al_2 O_3 – SiO_2]\) in terms of oxide constituents, hence the term triaxial porcelains [2].

The great interest in high strength of porcelain and the wide research on the porcelain system has resulted in three measure hypothesis which were described by Carty and Sennpati [3] as the mullet hypothesis, the matrix reinforcement hypothesis and the dispersion strengthening hypothesis respectively. According to Morita’s study’s replacing some portioning of feldspar with Barium Carbonate to prepare porcelainceramic, the replacement decreases the porcelain centering time[4].

Chaundhuri et al have been studied the mechanical and dielectric strength of porcelain ceramic which have been shown that each phase has its specific influence on these properties depending on its concentration and microstructural attributes[5].

Martin-Marquez et al studied the effect of firing temperature on sintering of porcelain. The sintering behavior of fired samples was evaluated by linear shrinkage, water absorption and porosity measurements. Both green powder and fired samples were characterized by means of differential thermal analysis (DTA), XRD, SEM and bending strength measurements[6].

Zahade et al studied the effect of alkaline oxide on the composition of porcelain. Firing shrinking, porosity, density and strength of the samples where studied, all them effecting by the addition of the alkaline oxide [7].

Sedghietal studied the effect of different fluxes on porcelain insulator properties and structure which shown that among used fluxes adding 1% barium carbonate increase the density and bending strength of porcelain [8].

Al – Bermany et al studied the influence of ZnO, ZrO_2 and TiO_2 on the mechanical properties of a porcelain body. They found that the addition of these materials made composite good medium for transferring ultrasound waves so they can be used as coated material to enhance the absorption coefficient of porcelain [9].

Olkode et al studied the effect of wood ash addition on the properties of porcelain. The flexural strength decreased at the increasing of wood ash content until 5% then the strength improved [10].

Almeida-Junior et al investigated the effect of extreme cooling methods on the flexural strength, reliability and shear bond strength of veneer porcelain for zirconia. They found
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that fast-cooling method showed a higher flexural strength but a lower reliability in porcelain under tension test configuration, where the slow cooling method decreased the shear bond strength [11].

Sui et al showed that the strength of zirconia-porcelain interface is derived from the mutual diffusion of zirconium and silicon at the interface. Also they found the great Vickers hardness and low toughness suggests the weakness of porcelain [12].

Chyad studied the fracture statistic of porcelain. Two parameters have been used SEM for this study the addition of ZrO₂ powder and effecting of sintering temperature [13].

Sedghi et al prepared six electrical porcelain compositions with different amount of alumina silica and fired at 1300°C. Density, porosity, bending strength and dialectical strength were measured. Increasing the amount of alumina up to 30% decreases quartz and cristobalite phases but increases corundum phase 3 to 5 times. SEM observation revealed that dense particles and uniform distribution of long and thin needle shaped mullite are predominant in sample microstructure with high dialectical strength [14].

Hariharan et al used Bagasse ash, which is the residue after the sugarcane juice extraction, as a partial replacement of porcelain body. The investigation reveals that high quality ceramic specimens could be achieved from blended material. Thus, sugarcane bagasse ash waste presents high potential for application in the manufacturing of ceramic products. Sample with 20% bagasse ash content resulted in lower porosity, water absorption and high dialectical strength compared to standard porcelain insulator [15].

Vudhivanich et al have studied the effect of zirconia content on Lucite crystallization and microstructural evaluation of porcelain ceramic–nanocomposite. The amount of ZrO₂ (0-65 wt.% addition has been found to be one of the keys factors controlling leucite formation and microstructure on the ZrO₂ surface on porcelain ceramic [16].

The employment of feldspar in the composition of ceramics has obvious effect on plasticity, transparency, thermal and mechanical properties. The feldspar plays a very important role in ceramic and glass industry. Substitution of feldspar can be utilized as a replacement. Finally due to the lack of feldspar and its substitutes in Iraq and the imported materials is usually expensive, the feldspar should be synthesized.

The aims of this work are to establish technological roots for the synthesis of feldspar substitute utilizing materials locally available and then synthesis the porcelain from these local materials with its characterization.

**Experimental Part:**

**Materials used:**
The materials used in this work were Iraqi Duekhla kaolin and Urhuma sand which brought from Geological Survey Company of the ministry of Industry, Baghdad, IRAQ. The chemical analysis of these materials was performed at the same company as shown in Table (1). The other materials are feldspar and PVA as a binder.

**Synthesis of feldspar**
Feldspar is not available in Iraq, so it can be synthesis as the following procedure:
- Commercial grade potassium carbonate {98%} was utilized while the other materials are kaolin and silica. The powders were mixed and milled via laboratory milling and sieving system. Powders below 75 micron used for subsequent steps. 20.12wt. % of K$_2$CO$_3$ mixed with 51.3wt. % of kaolin and 26.3wt. % of silica. After good mixing the powder was compacted at 13 MPa to increase the contact area of mixed components and accordingly to enhance solid-solid reaction. The compacted discs were heat treated up to 1150°C for two hours and 10 C/mln. For heating and cooling rate.

**Synthesis of Porcelain**

50wt% of kaolin, 25wt.% silica and 25wt.% of prepared feldspar are mixed and milled for two hours as mentioned before. 3% of PVA was added to the mixture as a binder. The result powder was compacted as discs (10x10mm) then sintered at (1200, 1250 and 1300°C) for two hours at 10 C/min. as heating and cooling rate.

**Characterizations**

**X-Ray Diffraction**

The feldspar powder was analyzed by X-Ray diffraction system type (XRD-6000, Shimadzu-Japan) operated at 35KV and 40mA using Cuka-radiation with wavelength of 1.54056 Å.

**Physical tests**

Bulk density and open porosity of the sintered samples were calculated by the water immersion Archimedes principle by using the related equations [17]:

\[
\text{Bulk density} = \frac{D}{(M-S)} \quad \text{(1)}
\]

\[
\text{Open porosity} = \frac{(M-D)}{(M-S)} \times 100 \quad \text{(2)}
\]

Where:

D is Dry weight (g), M saturated weight (g) and S is the weight of sample after immersing in distilled water and suspended in air through a balance (g).

2.2.3-Hardness test

The Vickers microhardness has been measured according to ASTM E (384-99) using the following equation:

\[
Hv = \frac{(1.8544P)}{d^2} \quad \text{(3)}
\]

Where, Hv is the Vickers hardness and P is the applied load and d is the indent diagonal. The weight used was 500 g.

**Dielectric properties**

Dielectric properties such dielectric constant, loss factor and loss tangent have been measured by the HP 4284A precision LCR meter system which used with range of (20Hz-5MHz). The system is computerized results.
Results and discussion

Fig. 1 represents the X-ray diffractogram of synthesized feldspar. This pattern shows sharper peaks and enhanced intensities. The d-spacing values better fit the ASTM slandered diffraction data files Vol. 7 Dec. 1997 of feldspar material [18]. The high angle peaks displayed clearly which is resembles other indication for good crystallinity for the synthesized feldspar.

Fig. 2 represents the effect of MgO content on the bulk density of electrical porcelain at different temperature. As shown in this figure as the content of MgO increasing the density is increased and that may be due the replacement of MgO particles in the main composition of porcelain assuming that the density of MgO is 3.85 g/cm$^2$ compared to that of porcelain. Also it is clear that the density of the composite increased with the sintering temperature having the highest value with 1350 °C, while fig. 3 shows the effect of MgO percentages on the porosity of porcelain at different temperature. The porosity is decreased sharply with the increasing of MgO with sintering temperature that may be due to the filling most of the open pores with MgO particles and making good dense body as seen with the result of density. Fig. 4 illustrates the behavior of Vickers' microhardness of porcelain with different percentage of MgO and different sintering temperature. The figure shows an increasing in the hardness. The highest value at (30%) of MgO and at 1350 °C and that may be due to a sufficient fusion for the ceramic body has been occurred which leads to an increase in the glass phase formation that decrease the amount of pores at the surface as shown that at the porosity and density results beside that may be due to the initiation of crystalline phase mullite which has a density of 3.17 g/cm$^2$ and corundum phase (alumina) which has a density of 3.98 g/cm$^3$. The presence of MgO particles in porcelain promote the development of a less viscose liquid phase, which is turn improves the densification kinetics as long as it is operated within defined liquefied the composition and this result agree with the work of Gigdemir et al (19).

Figs. 5-7 illustrate the effect of MgO content on the dielectric properties such dielectric constant and loss tangent and the dielectric loss factor at different temperatures. Fig. 5 shows that dielectric constant increased with the increasing of MgO content with higher temperature but this increasing is not too much may be due to the presence of some microcracks in the ceramic insulators, where these cracks are observed mainly near the quartz particles and those regions where the crystalline phase or mullite phase is less or absent. The nature of cracks in porcelain body is depending on the expansion coefficient of the matrix and the particles addition. Circumferential cracking than the matrix.

At higher temperature such 1350 °C, there will be a considerable amount of glassy phase, when there is a large amount of glassy phase present in the structure, the mobile of ions such Na$^+$, K$^+$, Al$^{3+}$ of the matrix and Mg$^{2+}$ of additives finds an easy path to more and hence increases the conductivity. On other hand, the hardness can be greatly increased by undisclosed quartz.

While Fig. 6 reveals the effect of MgO additive at different temperature on the loss tangent of the samples. As shown in the figure the loss tangent decreased sharply, again the lowest value of the loss tangent was at 1350 °C which decreased from 0.016 till 0.0043. The same behavior was for dielectric loss tangent as shown in Fig. 7.
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dielectric loss tangent decreased drastically specially at the lower MgO additive for all the temperatures and then decreased slowly.

Conclusions:
From the experimental results, it was concluding the followings:
1- Feldspar can synthesized from locally Iraqi raw materials
2- Electrical porcelain can synthesized from locally Iraqi raw materials.
3- Addition of MgO powder enhanced the physical and dielectric properties.
4- The higher values of density, hardness and dielectric constant at 30% MgO and sintering temperature of 1350°C.
5- The same percentages of MgO and sintering temperature reduced the porosity and loss tangent to lowest values.

Table (1) chemical analysis of Duekla kaolin and Urduma sand

| Oxides  | Kaolin | sand |
|---------|--------|------|
| SiO₂    | 47.26  | 98.4 |
| Al₂O₃   | 34.84  | 0.4  |
| Fe₂O₃   | 1.32   | 0.05 |
| TiO₂    | 1.4    | --   |
| CaO     | 0.15   | 0.3  |
| MgO     | 0.38   | 0.3  |
| Na₂O    | 0.25   | --   |
| K₂O     | 0.61   | --   |
| L.O.I   | 12.91  | 0.55 |

L.O.I : loss on ignition

Figure (1) X-ray defractogram of synthesis of feldspar showing the d – values
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Figure (2) Density of electrical porcelain against MgO additive

Figure (3) Porosity of electrical porcelain against MgO additive

Figure (4) Microhardness of electrical porcelain against MgO additive
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Figure (5) Dielectric constant of electrical porcelain against MgO content at 1MHz

Figure (6) Loss tangent of electrical porcelain against MgO content at different temperature at 1MHz

Figure (7) Dielectric loss factor of electrical porcelain against MgO content at 1MHz
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