

Dielectric Properties of Triaxial Porcelain Prepared Using Raw Native Materials Without Any Additions

S. Kitouni

Abstract— in this study, production of porcelain, for the ceramic dielectric applications by using inexpensive natural raw materials or waste materials was undertaken. The principal raw materials of porcelain, such as kaolin, feldspar, and quartz, are relatively inexpensive and readily available. The raw materials used in this study were collected from Algerian source (Kaolin was from Debagh deposit, Quartz from El Oued city and Feldspar from Ain Barbar deposit). The basic electro-porcelain composition was selected consisting of 37% kaolin, 35% potash-feldspar and 28% quartz. The samples synthesized were characterized by X-ray diffraction (XRD) technique.

The dielectric properties of porcelain bodies prepared without the incorporation of mineralisers or metal oxides, e.g. dielectric constant, dielectric loss tangent ($\tan \delta$) and loss factor, were investigated. Dielectric measurements have been carried out at 1 KHz in the temperature range ambient Temperature – 200°C. The dielectric constant ϵ' and dielectric loss tangent $\tan \delta$ respectively of porcelain samples sintered at the most proper sintering temperature 1200°C were about 21.22 and 0.006. The value of dielectric constant is higher as compared to conventional porcelain which not exceeds generally 9.

Index Terms— Dielectric constant, dielectric loss tangent, dielectric loss, feldspar, kaolin, quartz, porcelain,

I. INTRODUCTION

THE electric power industry tends to develop extra high voltage, large capacity and long distance transmission, the electric porcelain is required. An advantage of electric porcelain over other insulating ceramic materials is the fact that it enables one to make large size high-voltage insulators of a complex configuration. Electro porcelain can be classified as follows based on its purpose and properties: normal high-voltage porcelain used in the production of high-voltage line and apparatus insulators; high-voltage porcelain with an increased quartz content used to make high-voltage apparatus insulators with improved electromechanical properties; high-voltage alumina porcelain for high-voltage apparatus insulators with elevated mechanical strength; low-voltage porcelain used in the production of insulators and insulating parts for plants up to 500 V generating direct and alternating current and weak current plants [1]. It has been estimated that more than 20% of the total outlay for a typical transmission

and/or distribution system of electric energy is spent on insulation alone and prominent among them is porcelain [2]. The utilization of ceramic materials as electrical insulators goes back until 1850 when Werner von Siemens introduced in the construction of electrical air lines the use of electrotechnical porcelains. During this long period of time it has been realised that several characteristic properties of porcelain (e.g. mechanical strength, high-power dielectric strength and corrosion resistance) as a ceramic product cannot be obtained in other materials. Today, the growing demand for porcelain in the field of electrical engineering, caused by the importance of electric energy in modern society, motivates many research projects in order to obtain the best properties for the requirements and applications of porcelain insulators [3]. Basically porcelain is an insulator but the rise of temperature as example may damage its insulation property due to decrease in its resistivity and the porcelain body may be employed as conductor [4].

Electrotechnical porcelain is a clever compromise between electrical, thermal and mechanical resistances. Improving performance characteristics of the electrical porcelain involves the updating of its production technology. The characteristics of porcelain insulators depend to a marked degree on the percentage composition of the mixture, sources of the mixture and method of manufacture. The production of electrotechnical porcelain is based predominately on natural raw materials. To this end, kaolin-feldspar-quartz triaxial porcelain was prepared from locally available raw materials and characterized. The characterisation is aimed at comparing the electrical properties of the triaxial porcelain with those of others works and hence explores the possibility of local substitution of the imported variety which is the subject of a future research.

II. EXPERIMENTAL PROCEDURE

A. Raw materials

The raw materials used in the preparation of the triaxial electrical porcelain are kaolin, feldspar, and quartz (in the form of silica sand). The raw minerals used in the present investigation were taken from different deposits in Algeria. The sources of the raw materials and their chemical composition are shown in table 1. This study investigates a

S. KITOUNI, is with the Physics Department, Constantine 1 University, Constantine, 25000, ALGERIE. (e-mail:souad0714@yahoo.fr).

unique porcelain type percentage composition of Kaolin, 37: feldspar, 35: and quartz, 28.

B. Apparatus and Procedure

We attempt to make samples of electro technical porcelain from the local raw materials. The technological process of electro porcelain production consisted of several stages: preparation, combination, and mixing of the initial raw materials in the required proportions; formation of an article with the required shape from powder (mix); heat-treatment (sintering). The samples obtained consisted of 13 mm in diameter cylinders. The samples were prepared by the dry process technique and fired at a temperature of 1200°C with a soaking time of 2 hours for proper sintering. To avoid spurious effects and in order to create a conduction path for electrical measurements on the samples, samples were electroded with silver paste on the top and bottom faces. Electrodes were fired at 300 °C for 20 mn with a heating rate of 5 °C/min. All measurements were undertaken at range temperature 20-190°C on a LCR meter connected to a furnace heating rate of 5 °C/min) as shown in Fig. 1. Effective permittivity was calculated as measured capacitance multiplied by the geometrical factor of a sample. The dielectric constant was determined from the relation where C_0 , is the capacitance of a two-electrode system with air as the dielectric, and C is the capacitance with the sample as a dielectric.

$$\epsilon' = \frac{C}{C_0} = \frac{Cl}{\epsilon_0 A} \quad (1)$$

$$\epsilon'' = \epsilon' \tan \delta \quad (2)$$

Where l is the thickness of the sample, A is the effective area between the electrodes and ϵ_0 is the free space permittivity. The dielectric loss tangent ($\tan\delta$) was measured directly from the instrument.

TABLE I
THE SOURCES AND CHEMICAL COMPOSITION OF THE RAW MATERIALS

Mineralogical Name	chemical composition	Source
Kaolin	$Al_2O_3 \cdot 2SiO_2 \cdot 2H_2O$	Debagh deposit
Feldspar	$K_2O \cdot Al_2O_3 \cdot 6SiO_2$	Ain Barbar deposit
Quartz	SiO_2	El Oued

III. RESULTS

The dielectric constant (ϵ') of the sintered bodies as a function temperature measured at 1 KHz is shown in Fig. 2. The dielectric constant remains fairly constant in the ambient - ~100 °C, thereafter increases gradually. The dielectric constant is almost constant at low temperatures and the dielectric constant dependence increases with increasing temperature. It is interesting to note that as the temperature increases the dielectric constant increases. The increase in dielectric constant temperature is one of the features of normal

dielectrics. The rise in ϵ' is ascribed to the dc conductivity effect [5]. The electrical properties of the samples were determined beforehand. The room-temperature permittivity at 1 KHz is 21.22. Based on a mixture rule using various theories to predict the dielectric constants of materials, the dielectric constant of the composites was found to be 12~24. Prediction of the dielectric constants is considered on the basis of phases, shape of phases, and porosity [6].

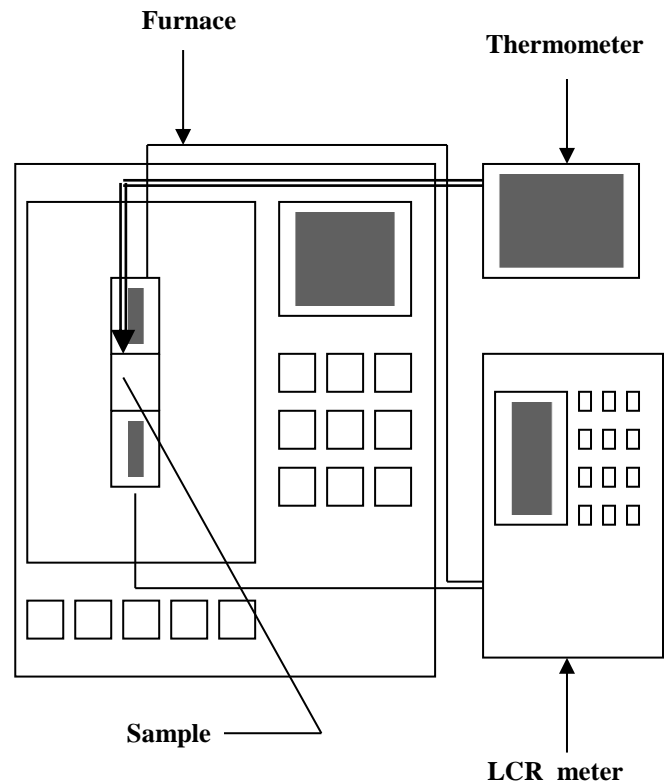


Fig. 1 Test setup for capacitance and dissipation factor

An increase of dielectric constant with increasing temperature results from polarization phenomena at grain boundaries and electrodes [7]. In ceramic materials, ions can be carriers to provide electrical conduction. The degree of conductivity is largely dependent on the energy barrier that must be overcome for the ion to move from one lattice position to the next. At low temperature, conductivity is low. However, if the temperature is high enough to overcome the barrier for lattice diffusion, the conductivity increases.

Other reason which can be given is some form of interactions between the dielectrics and Ag electrodes are responsible for the higher ϵ' . Moreover, the lattice defects formed could accelerate the mass transport process; the existence of lattice defects such as vacancies and interstitials in the structure aids conduction [5], further promoting grain growth. As is well known, the grain size will influence the dielectric constant [8]. Thus the insulator could be used, as capacitor bushings where low charge storage capacity is very desirable.

Dielectrics with dielectric constant above 12 are generally materials for capacitors and transducers. The ceramic capacitors are one of the most widely used discrete electronic

components that play a very important role in electronic industry. In the recent years, a rapid fast development in the ceramic capacitor technology has been achieved to meet the needs of advancement in microelectronics and communication. It is envisaged that more applications of ceramic capacitors will be found in future [8]. The porcelain sample is suitable for this application in the present form because of the higher dielectric.

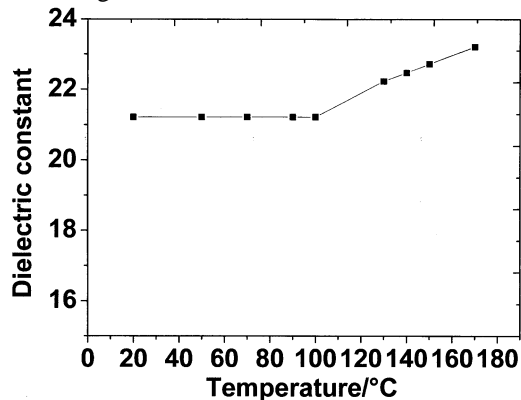


Fig. 2 Temperature versus dielectric constant for samples sintered at 1200 °C for 2 h

The dielectric loss factor (ϵ'') of the sintered bodies as a function temperature measured at 1 KHz is shown in Fig. 3. A similar trend is observed with ϵ' .

The increase of ϵ'' with temperature can be explained by the relaxation phenomenon which is divided into three parts, conduction losses, dipole losses and vibration losses [9].

At low temperatures conduction losses have minimum value. As the temperature increases ϵ'' conductivity increases and so the conduction losses increase. As a result increase the value of ϵ'' with increasing temperature.

The dielectric loss tangent ($\tan \delta$) of the sintered bodies as a function temperature measured at 1 KHz is shown in Fig. 4. Variation of dielectric loss tangent ($\tan \delta$) as a function temperature at 1 KHz the samples under study is consistent with the ϵ' behaviour as shown in Fig. 2 of the corresponding samples.

The room-temperature dielectric loss tangent is 0.006. In the dielectric materials, defects, space charge formation, lattice distortions, etc., in the boundaries could produce absorption current resulting in $\tan \delta$ [10]. The dielectric loss tangent and dielectric constant of samples increase with increasing temperature. The electron part of polarization in an ionic crystal decreases with increasing temperature due to decreasing density of the material, and the ionic part of polarization is affected by two opposite factors: decreasing density and weakening of elastic bonds, which cause a decrease in the elastic bond coefficient. The dielectric constant of ionic cubic crystals with increasing temperature grows relatively little and is nearly linearly [6].

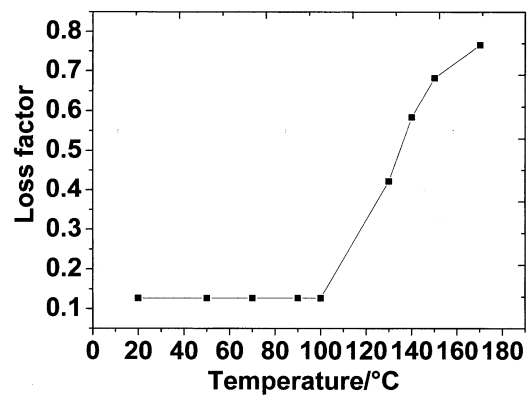


Fig. 3 Temperature versus loss factor for samples sintered at 1200 °C for 2 h

The porcelain body is composed of phases such as mullite, quartz and glass. Its electrical properties are dependent on the properties of each of these phases, both at room and at elevated temperatures. Besides, microstructural characteristics, such as crystal size and size-distribution also contribute to the electrical properties of the body because they influence the passage of current through the body. X-ray structural analysis (DRX) showed the presence of mullite, separated quartz and a glassy phase in the samples. The DRX was elaborated in an earlier paper [11]. In this study, the glassy phase is derived from the potash feldspar and quartz component in the porcelain composition. Glass phase has a dominant influence on electrical and dielectric properties of fired ceramics. These properties are determined by the concentration and mobility of K^+ and/or Na^+ ions in this phase [7]. In the presence of the large amount of glassy phase in the structure [11], the mobile ions such as K^+ finds an easy path to move and hence increases the conductivity [5]. It is known that the dielectric constant of porcelain increases in presence K^+ and Na^+ cations and decreases when they are replaced by Ca^{2+} , Mg^{2+} , and Ba^{2+} cations [12]. On the other hand mullite, which is a crystalline phase, has a vital role on electrical properties. As the matrix of the ceramic sample is clay matrix or the glassy phase, mullite maintain the stress level in a higher order as just in a composite matrix [13].

In addition mullite behaves like semiconductor at high temperature. The band gap energy (E_g) of mullite is 1.43 eV and this value is in the semiconductor interval [12]. Mullite crystal has inherent defect centres as O^{2-} ion vacancy in its structure and it is a potential ionic conductor.

Substitution of Al^{3+} ion in the (AlO_6) octahedral and (AlO_4) tetrahedral sites of mullite lattice by the transition metal ions, Mn^{n+} can make it n- type (if $n > 3$) and p-type (if $n < 3$) electronic conductor.

Apart from this, substitution of Al^{3+} ion by M^{n+} ion (if $n=3$) in the octahedral or tetrahedral position and only incorporation of M^{n+} ion in the interstitials or structural channels of mullite lattice can impart electrical conductivity in mullite [4]. In this study, the dielectric constant obtained in the present study is higher than the value for pure mullite (6.9) [14].

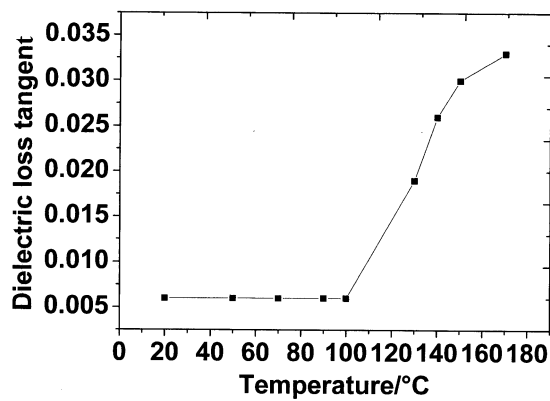


Fig. 4 Temperature versus dielectric loss tangent for samples sintered at 1200 °C for 2 h

IV. CONCLUSION

The present work is an attempt for the elaboration of aluminosilicate porcelain samples with simple techniques. The data obtained show that Local raw materials are quite suitable for fabricating electro technical porcelain. Based on the experimental results obtained in this study, using the locally available raw materials, electrical porcelain with good dielectric properties can be produced. This material can be used for dielectrics in commercial capacitor applications. Thus the insulator could be used, as capacitor bushings where low charge storage capacity is very desirable. The porcelain sample is suitable for this application in the present form because of the higher dielectric.

ACKNOWLEDGMENT

This study was conducted in the ceramics laboratory of physics department (Constantine 1 University, Constantine, Algeria).

REFERENCES

- [1] Z. V. Kolpashchikova, E. V. Shcherbakova, N. S. Kostyukov, "Polarization processes in electrotechnical porcelain within a wide frequency range", *Glass and Ceram.*, vol. 60, no.11-12, pp.370-373, 2003.
- [2] L.U ANIH, "Characterization of Locally Sourced Kaolin-Feldspar-Quartz Triaxial Porcelain for Insulation Applications", *Electric Power Engineering Conference (EPEC)*, June 2005.
- [3] J.M.Amigo, J.V. Clause, E. Vicente, J. M. Delgado, M. M. Revento, L. E. Ochando, T. Debaerdemaeker, F. Marti, "X-ray powder diffraction phase analysis and thermomechanical properties of silica and alumina porcelains", *J. of the Eur. Ceram. Soc.*, vol. 24, no.1, pp.75-81, 2004.
- [4] S.P. Chaudhuri, P. Sarkar, A.K. Chakraborty, "Electrical resistivity of porcelain in relation to constitution", *Ceram. Inte.* vol. 25, no.1 pp.91-99, 1999.
- [5] A. S. Demirkiran, R. Artir, E. Avci, "Electrical resistivity of porcelain bodies with natural zeolite addition", *Ceram. Inter.* vol. 36, no.3, pp.917-921, 2010.
- [6] S.J. Hwang, Y.J. Kim, H.S. Kim, "La₂O₃-B₂O₃-TiO₂ Glass/BaO-Nd₂O₃-TiO₂ ceramic for high quality factor low temperature co-fired ceramic dielectric", *J Electro. Ceram.*, vol. 18, no.1-2, pp.121-128, 2007.

- [7] V. Tmova, I. Fura, F. Hanic, "Influence of technological texture on electrical properties of industrial ceramics", *J. of Phys. and Chem. of Sol.* vol.68, no.5-6, pp.1135-1139, 2007.
- [8] Ying-Chieh Lee, "Dielectric Properties and Reliability of Zn_{0.95}Mg_{0.05}TiO₃+0.25TiO₂ MLCCs with Different Pd/Ag Ratios of Electrodes", *Int. J. Appl. Ceram. Technol.*, vol. 7, no.1, pp.71-80, 2010.
- [9] H. M. El-Malah, N. A. Hegab, "Studies on a.c. properties of Ca_{1-x}Sr_xTiO₃ perovskites", *J. Mater. Sci.* vol.42, no.1, pp.332-336, 2007.
- [10] G.B. Kumar, K. Sivaiah, S. Buddhudu, "Synthesis and characterization of ZnWO₄ ceramic powder", *Ceram. Inter.* vol.36, no.1, pp.199-202, 2010.
- [11] S.Kitouni, A. Harabi, "Sintering and mechanical properties of porcelains prepared from Algerian raw materials", *Cerâmica*, vol.57, no.344, pp.453-460, 2011.
- [12] V.P. Il'ina, "Feldspar material from Karela for electrical engineering", *Glass and Ceram.*, vol.61, no.5-6, pp.195-197, 2004
- [13] R.A. Islam, Y.C. Chana, M. F. Islam, "Structure-property relationship in high-tension ceramic insulator fired at high temperature", *Mat. Sci. and Eng.* vol.B106, no.2, pp.132-140, 2004.
- [14] V. Viswabaskaran, F.D. Gnanam, M. Balasubramanian, "Mullite from clay-reactive alumina for insulating substrate application", *Applied Clay Sci.* vol.25, no.1-2, pp.29-35, 2004.

BIOGRAPHIES

S. KITOUNI received Ph.D. degree in physics from the University of Constantine, Algeria in 2013. Her current research interests are dielectrics. Participate in some international conferences in Algeria.