The Effects of Frequency and Temperature on the Dielectric Constant of $X\%Al_2O_3$ Powder Extracted From Iraqi Kaolin

A S Karim
College of Science Education
University of Sallahadin

Abstract

Samples of $X\%Al_2O_3$ (X = 17.85, 28.05, 87.98 and 95.63) were prepared from Iraqi kaolin, after calcinations process, by the method of evaporation. From the temperature dependence of capacitance, the temperature dependence of dielectric constant are studied for the samplesof annealed powders of various Al_2O_3 contents in the frequency range 1KHz – 1MHz.

The frequency dependent values of dielectric constant for the powder content 87.98% Al_2O_3 and 95.63%Al_2O_3 are found to be varies between 9.21 – 12.29 and 6.99 – 12.063, respectively. While for the others percentage of 17.85%Al_2O_3 and 28.05%Al_2O_3 powder content samples, it changes between 22.195 – 151 and 13.015 – 21.09 , respectively. These dependence are analyzed and discussed in details.

The annealing temperature effect on the dielectric constant are given for the frequency investigation of 10KHz. However, the annealing temperature transforms the structure and that consequently affects the dielectric constant. The dielectric constant found to be decreases with increasing the temperature for all the investigated samples from 17.85%Al_2O_3 to the 95.63%Al_2O_3.

Keywords : Dielectric constant, Al_2O_3 powder, annealing temperature

تاثير التردد والحرارة على ثابت عزل مسحوق $\%Al_2O_3$ Xالمستخرج من كاؤولين العراقي

أيوب صابر كريم

كلية تربية العلوم – جامعة صلاح الدين

المستخلص

تم تحضير نماذج من $X\%Al_2O_3$ بنسب ($X = 17.85, 28.05, 87.98$ and 95.63) من كاؤولين العراقي بعد عملية التحميص و بطريقة التبخير . من الاعتماد الحراري للسعة ، تم دراسة ثابت العزل للمسحوق المحمص لمختلف النسب من $x\%\ Al_2O_3$ و تحت تأثير التردد في المدى 1KHz - 1MHz .

وجدت بأن الاعتماد الحراري لثابت العزل للمسحوق التي تحتوي على نسب $\%Al_2O_3$ 87.98و $95.63\%\ Al_2O_3$ هي 12.29 - 9.21 و 12.65 - 6.99 على التوالي . بينما لبقية النماذج التي تحتوي على نسب $١٧.٨٥\%Al_2O_3$ و Al_2O_3 ٢٨.٠٥% فانها تتغير بين 151− 22.195 و 21.09 - 13.015 على التوالي. تم تحليل ومناقشة هذه الاعتمادية بالتفصيل.

كذلك تم دراسة تأثير عملية تحميص على ثابت العزل عند التردد 10KHz . من تاثير حرارة التحميص وجدت بأنّ التركيب البلّوري هي السبب الاساسي لتقليل قيمة ثابت العزل عندما للنماذج $17.85\%\ Al_2O_3$ الى $95.63\ \%\ Al_2O_3$.

Introduction

Ceramic suitable for use in capacitors are characterized by a high dielectric constant. In conventional ceramics, however, the dielectric constant has shown a strong dependence on temperature. The behavior of the capacitance with respect to temperature is quantified in a single term called the temperature coefficient of capacitance (hereafter referred to as TCC which is expressed in terms of parts per million (ppm/degree.C). Careful material selection and processing have made possible the development of dielectric ceramic bodies that have very small TCC's over a wide range of temperature. This phenomenon can occur when a multiphase body is formed in which each phase with a positive TCC is counterbalanced by one or more phases with an equivalently negative TCC. Certain bodies in which this compensating effect is present have temperature coefficient of capacitance that are usually small and are known as Negative-positive-Zero or NPO bodies [Inventors, 1990] .

The demand for ceramics in the electrical industry has phenomenally both in quantity and quality during last and this century. Singer and Singer 1963 and Budnikov 1964, classified ceramic materials for electrical industry and their general properties. Graham and Tallen 1971 briefly classified and described the broad field of polycrystalline electrical insulators. Ceramic materials that satisfy the value of dielectric constant and dielectric loss index at 1 MHz, (≤ 30 and ≤ 0.03), respectively at 25 ºC , would be classified as a good electrical insulators [Buchman, 1981 and Van vlack, 1964].

In the last fifty year, a great deal of assessment has been carried out in manufacturing electrical insulators. Bauer and Dietz, 1981, discussed the future needs for composite and porcelain electrical insulators. Other workers [Nortan, 1978; leda, and tanaka, 1986; and Roggen, 1990] outlined in some detail the development, which took place in the instrumentation and methods of studying such insulators.

The dielectric constants of comparably grown unpatterned films of Al_2O_3 has been measured and found to be 8.9 - 9 at various film thickness and measurement temperatures [Biercuk et al. 2003] .

Alumina is an important electro ceramic material with ultra-low dielectric constant ($\varepsilon_{r \sim 9.8}$). Many papers have been published to report the origins of dielectric loss, which fall into two categories: intrinsic loss is dependent on the crystal structure and can be described by the interaction of the phonon with the ac electric field. The ac electric field alters the equilibrium of the phonon system and the subsequent relation is associated with the energy dissipation. Extrinsic losses are associated with imperfections in crystal structure, e.g., lattice disorder, point defects, dislocations, grain boundaries, random crystalline orientation, impurities, porosity and microcracks. Alford et al. have made a profound investigation of the extrinsic loss of polycrystalline alumina [Song et all.; 2007].

Rasin, F.A. in (1998) has been studied the dielectric properties for types of material rocks. The rocks were porcelanite, the major mineral, that constitute it, is OPAL-CT and the second is Ninivite rocks, the major mineral, that constitute it, is quartz. The dielectric constant of ceramic body is affected by changes in its composition, manufacturing technique and density (porosity) as well as by testing factors including the effect of temperature and voltage frequency [Frank, 1962; Harrop, 1972].

In this work temperature dependence of capacitance, and factors affects $X\%Al_2O_3$ capacitance are investigated. The effect of calcinations on dielectric properties of powder extracted from Iraqi kaolin will also be studied.. In addition, dielectric constant of $X\%Al_2O_3$ with various powder contents at different frequency region will be discussed.

Experiments

A- $X\%Al_2O_3$ materials and formation process

$X\%Al_2O_3$ was chosen as a ceramic powder with various ratios , at various of X (X= 17.85, 28.05, 87.98 and 95.63), because it is one of the widely-used high dielectric constant powders. In this work, the Al_2O_3 powder with various contents of X, produced by evaporation method, from Iraqi Kaolin, which is an abundant material in Iraq. The agent solvent H_2SO_4 was used for materials extraction [Karim, 2006]. Samples were provided with 13mm diameter discs and 0.3822mm of average thickness.

B- Capacitor fabrication and characterization

A schematic view of the measurement technique is shown in Fig.(1) . Two parallel conductor, 12.002 mm in diameter was used to fabricate this capacitor so as one of them is fixed and the other is movable, their adjustment were controlled with a small coiled spring to allowed both the sample and the system expansion and also contraction throughout both heating and cooling. A two coiled short circuit heaters made from a copper wire with a diameter of about (0.5mm), with a use of (1.45-1.55) DC applied voltage.

To prevent the effects of the electromagnetic waves the heaters and wires used in the circuit were covered by a thin aluminum foil. Capacitance measurements C were performed by a programmable RCL meter at various frequencies in the temperature range (27-110) ^{o}C. The capacitor system was placed inside a thermally insulated cavity made of cork materials.

The test specimen and guard electrode are designed in such away that the sample diameter be larger than that of guarded electrode by at least 0.994mm. In these conditions the field distribution in the guarded area will be identical with that existing when vacuum is the dielectric, and the ratio between these two direct capacitance is the dielectric constant. One advantage of the RCL meter method is that, since the powder samples are in contact with metal electrodes, it is free from any influence of both the electrodes and the electromagnetic effects on Alumina transition properties. The RLC digital starts by arranging the earth contacts for the capacitance to discharge any of the existent electromagnetic wave, throughout the measurements. The measuring frequency range were from 1KHz to 1MHz.

C- Accuracy, Precision and Calibration Concepts:

The thickness must be determined by measurements distributed systematically over the area of the specimen that is used in the electrical measurement. In the present work the thickness was measured using micrometer with an accuracy of ± 0.02mm and the average value is taken into account. In this field, one could take into account, that error calculation follows the rules defined by the American Nation Bureau of standards [Rasin, 1998]

Copper-constantan as thermocouple devices were chosen for the measure of samples temperature in the range of (27 - 110) ºC. The copper constant thermocouple was calibrated by comparison the data readed from digital thermometer with that of the mercury thermometer where both are inside of a heated water and the detailed are given else where [Abbas, 2005] . The LCR- meter was calibrated using a built-in calibration software, followed by using standard capacitors and resistors. The sample handling precaution are in accordance with ASTMD 1371, [Rasin, 1998]. The measuring frequency range is sufficient to satisfy the requirement for dielectric measurements in the present work.

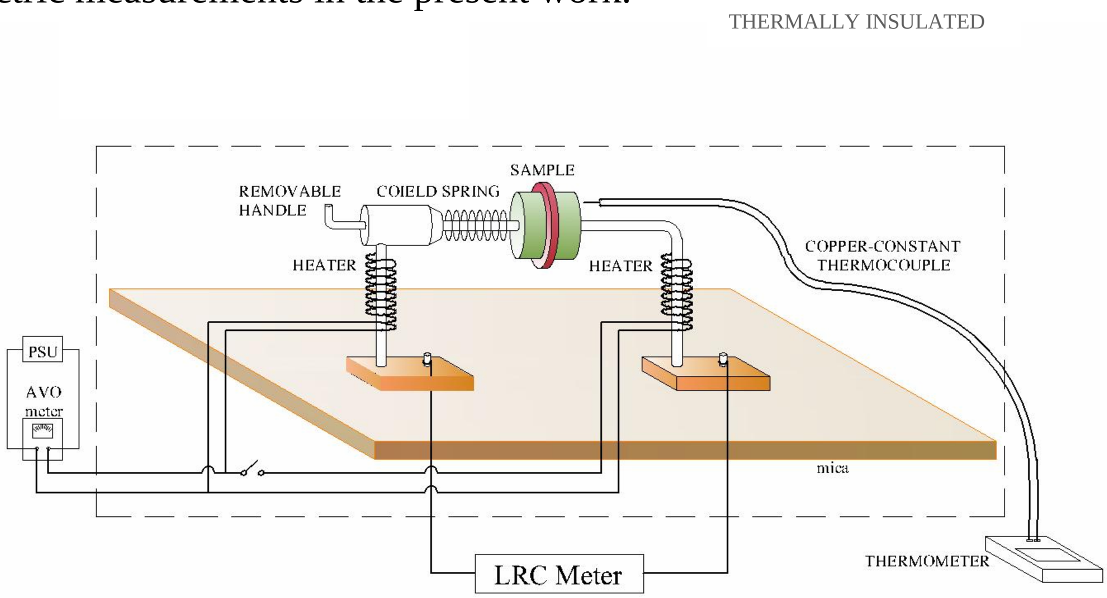

Fig: (1) : Schematic diagram of the capacitor system and dielectric measurment.

Temperature dependence of $X\%Al_2O_3$ capacitance

Generally, capacitance of capacitors can be expressed as

$$C = \varepsilon \frac{A}{t} = \varepsilon_o k \frac{A}{t}, \quad \left(k = \frac{\varepsilon}{\varepsilon_o} \right), \quad \ldots\ldots\ldots\ldots (1)$$

C is capacitance, k is relative dielectric constant, ε_o and ε are vacuum and dielectric material's permittivity, t and A are the thickness and area of a dielectric material. Based on the above equation, the relation between temperature dependence of capacitance and dielectric constant can be expressed as [Hyun et al., 2005]

$$\frac{1}{C} \cdot \frac{dC}{dT} = \frac{A}{t} \frac{1}{k} \cdot \frac{dk}{dT} \ldots\ldots\ldots\ldots (2)$$

Where $\left\{ \frac{1}{C} \cdot \frac{dC}{dt} \right\}$ is defined as temperature coefficient of capacitance (TCC) and $\left\{ \frac{A}{t} \frac{1}{k} \frac{dk}{dT} \right\}$ is defined a temperature dependence of dielectric constant (TCK). Because thermal expansion coefficient of metal electrodes is in the range of ($\sim 10^{-6}$/K) and the dimension of the metal electrode increase uniformly as the sample temperature increases it is possible to assume that the temperature dependence of electrode area A and thermal expansion coefficient of Al_2O_3 samples thickness ($\sim 10^{-6}$/K) are neglible [SAINT, 2005]. However, powder contain Al_2O_3 annealed at temperatures higher than 80^o C and then pressing under a pressure of about 10 tone will produse a sintering form material where it was originally in a pores state. As a result, the porosity is so much lowest for the samples so as their effects also could be neglected, then, the temperature dependence of capacitance of Al_2O_3 is defined by

$$(TCC) = (TCK) \ldots\ldots\ldots\ldots (3)$$

Therefore, the main factor of temperature dependence of $x\%Al_2O_3$ capacitance is determined by changes of dielectric constant of $x\%Al_2O_3$ s only. Figs. 2(a), (3a), (4a), (5a) show temperature dependence of capacitance. From these values, dielectric constants for $X\%Al_2O_3$ composites were calculated and plotted as shown in Figs. 2(b), 3(b), 4(b), 5(b).

From Fig.2(a)and (b), show the temperature dependence for both capacitance and dielectric constant of 17.85%Al_2O_3 .Their values decreases in the range from 30 ºC to 60 ºC and increased in between 90 to 110 ºC for both 1KHz and 10KHz .The reason for this could be due to the dielectric constant at low frequencies which can be attributed to molecular polarization of the asymmetric pyridine group in $Al_2(SO_4)_3.18H_2O$ [Karim, 2006]. At high frequencies since these groups can not maintain the alignment in the alternating field [Wyllie, 1960] then only electronic polarization exists. Calcinations of sample's at 600ºC, causes to a removal of hydrated polarization, then both capacitance and dielectric constant remains slightly changeable throughout the investigated temperature range (30-110 ºC) as indicated in Figs.3(a), (b). In which however, both of the dependence affected strongly by the applied frequency and they are increase systematically with temperature. It is clear from the two Figures of 2 and 3, the increase of Al_2O_3 content will make both the temperature and frequency depend for both capacitance and dielectric constant in a systematical form. However, the occurrence of the dehydroxylation process at 600ºC may probably be the reason. The explanation is the formation of $Al_2(SO_4)_3$ which a weight loss signals break-up. The dependence of loss factor, values with temperature and for three different frequencies are given in Figures 2c and.3c [Karim, 2006].

Fig. 3(b) shows the variation of dielectric constant (ε_r) with temperature for 28.05%Al_2O_3 at different frequencies. Dielectric constant increase slightly with temperature at 10KHz, 100KHz, while at 1MHz the amount of increasing approximately changes more than that occures for 10KHz, 100KHz. At high frequencies, since the groups can not maintain the alignment in alternating field, only electronic polarization exist. Similar results however , been observed in the case of ($Al_5Y_3O_2$) glass ceramics and other conventional number ceramic materials [Sutton, 1989]

Fig.(4) shows changes the capacitance, dielectric constant and loss factor tanδ vs. temperature at various frequencies for the 87.98% Al_2O_3 powder content. As the temperature elevated up to 60 ºC, both capacitance and dielectric constant decreased while loss factor increases. Above 60 ºC, the change rates started to remain constant. This may be due to maintaining phase transition for gamma-Al_2O_3 powder [Alford, 1996]. A higher amount of capacitance and dielectric constant was also observed for 1MHz in comparison to that of the frequencies of 10KHz and 100KHz.

Fig.5 shows the change of capacitance , dielectric constant and loss factor vs. temperature for 95.63% Al_2O_3 power contents at various frequencies. For this constinent, the capacitance, dielectric constants and loss factor are changes very slightly through the temperature range investigated. However, the crystal phase structure in this case remain as alpha-Al_2O_3 throughout all the temperature range investigated.

The dielectric constant and loss factor dependence on the applied frequency under the effect of annealing temperatures were also investigated. The curve shape in both Figures of 6 and 7 are indicated the increase of molecular polarization contributes, especially in the frequencies range of 10 – 200 KHz [Rasin, 1998].

Fig**.** 8 and Fig.10 shows dependence of dielectric constant with the annealing temperatures as well as with that of Al_2O_3 percentage powder content samples, respectively. As shown from these two indicated figures , the dielectric constant for the as-synthesised sample, dried at 80 ºC for 24h of 17.85%Al_2O_3 and for annealing at 600 ºC of 28.05%Al_2O_3 at10KHz are (151.559) and (13.015), respectively. This dielectric constant reduction is also observed for 87.98% Al_2O_3 and 95.63%Al_2O_3 powder contents. The dielectric constant for these two powder contents at the annealing temperatures1000 and 1200ºC and at the frequency of 10 KHz are (11.045) and (6.995), respectively. The effective dielectric constant of X%Al_2O_3 powder content is linearly proportional in the temperature range from 80 ºC to the 1000 ºC for values of X from 17.85%Al_2O_3 to 87.98% Al_2O_3 . Above this range, the relaxation expected to increase more strongly which give a more dielectric relaxation. Therefore the dielectric constant reduction of 87.98% Al_2O_3 and of 95.63%Al_2O_3 is mainly expected to be due to a phase transformation from gamma alumina at 1000 ºC to that of alpha alumina at 1200 ºC [Karim, 2006]. However, the above explanation can also be used to explain the loss factor tanδ, given in Fig.9.

However, the dielectric constant of 87.98% Al_2O_3 and 95.63%Al_2O_3 powder contents varies between 9.21 – 12.29 and 6.99 – 12.063, respectively, at the range of frequencies 10KHz, 100KHz and 1MHz at 30 ºC, as shown in Figs. 4(b) and 5(b) and Tab.1 . However, at the same range of frequencies for the other percentages 17.85%Al_2O_3 and 28.08%Al_2O_3 powder samples its change between 22.195 – 151 and 13.015 – 21.09, respectively, that is due to the dominates of $Al_2(SO_3).16H_2O$ and $Al_2(SO_3)$ [ayoub, 2006].While for the first two

Al_2O_3 percentage samples the phase structures are γ and α- alumina [ayoub, 2006].

Table1: Dielectric constant for $X\%Al_2O_3$ various frequencies at room temperature.

Annealing temp.	$X\%Al_2O_3$ 17.85	10kHz	100 kHz	1MHz
as synthesised		151.5	22.1955	33.519
600 ºC	28.05	13.0152	13.7856	21.0965
1000 ºC	87.98	11.0454	9.21157	12.2977
1200 ºC	95.63	6.99586	10.5375	12.063869

Conclusion

The alumina extracted from Iraqi Kaolin by evaporation method, at various Al_2O_3 powder contents, are suitable to use as an insulator ceramic material. Annealing temperature is controls the amount of dielectric constant as well as the loss factor of $X\%Al_2O_3$ content ceramics . The material also suitable to use as a high and low frequency capacitance.

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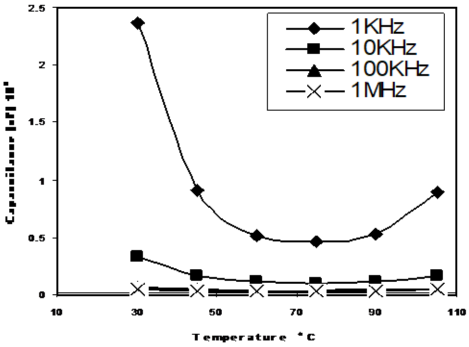
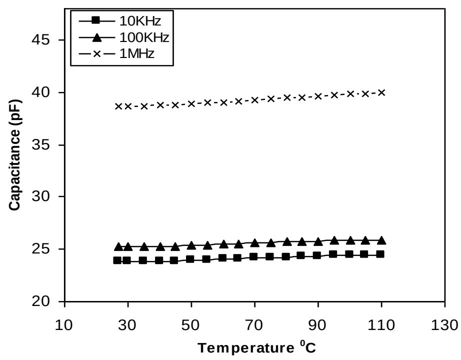


(a)



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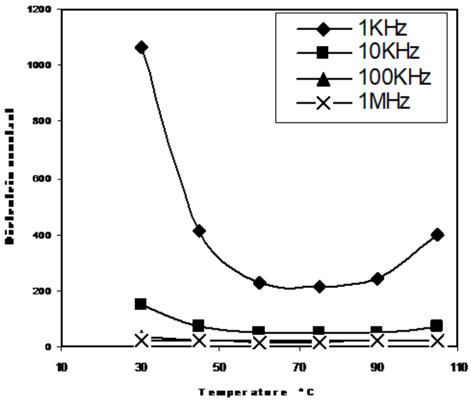

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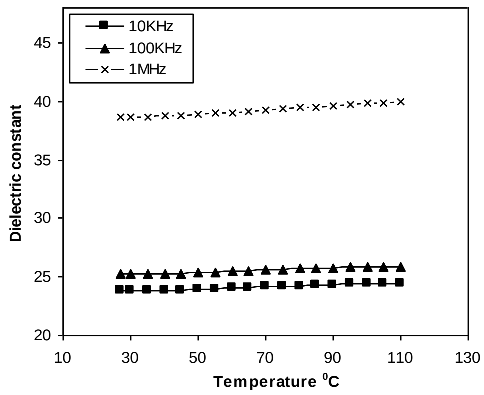

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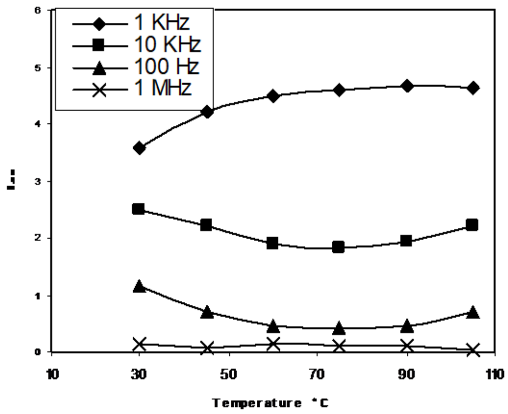

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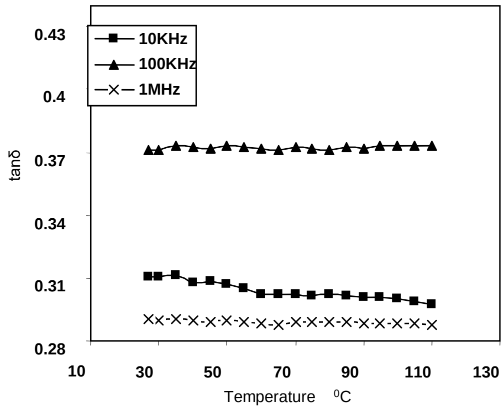
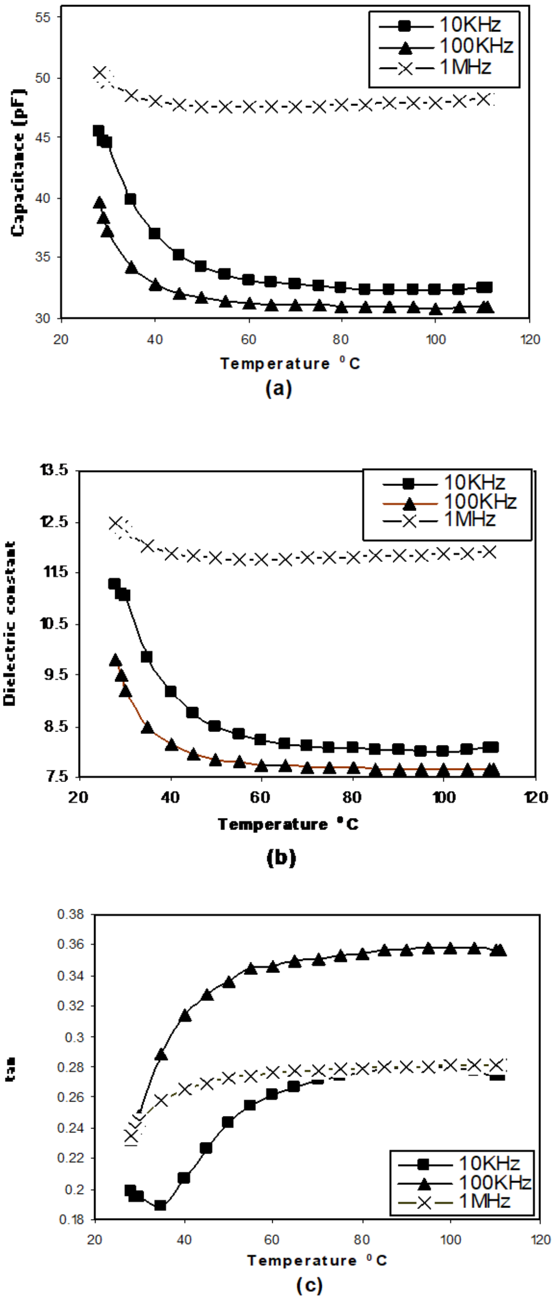


(c)

Fig.2 Variation of (a) C

Fig.3 Variation of (a) capacitance, (b) dielectric constant, and (c) loss factor vs. temperature for constant, and (c) loss factor vs temperature for 17.85%Al_2O_3 powder contents. 28.05%Al_2O_3 powder contents.



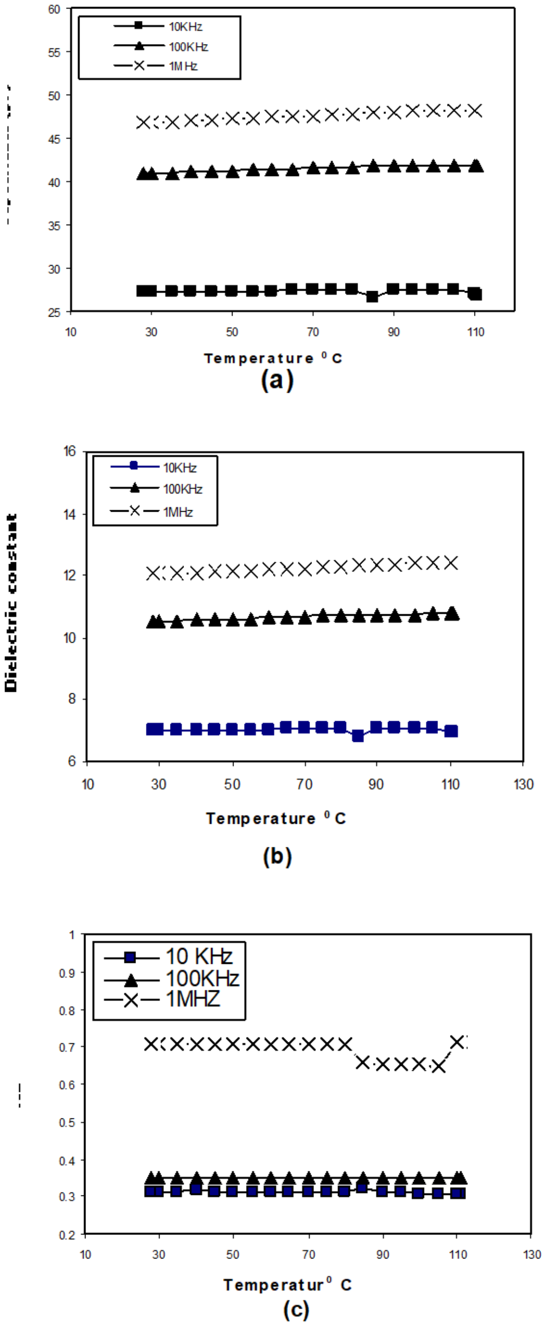


Fig.4 Variation of (a) Capacitance, (b) dielectric Fig.5 Variation of (a) Capacitance, (b) dielectric constant, and (c) loss factor vs. temperature for constant, and (c) loss factor vs. temperature for 87.98%Al_2O_3 powder contents. 95.63%Al_2O_3 powder contents.

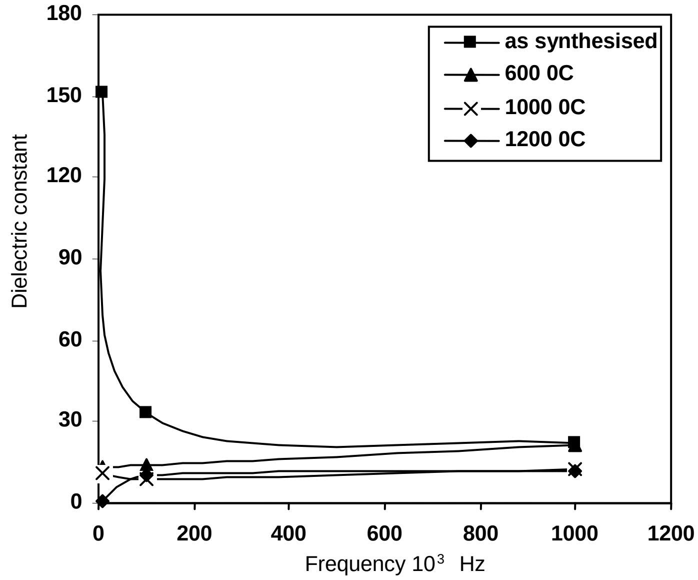


Fig.6 dielectric constant versus frequency for various annealing temperatures

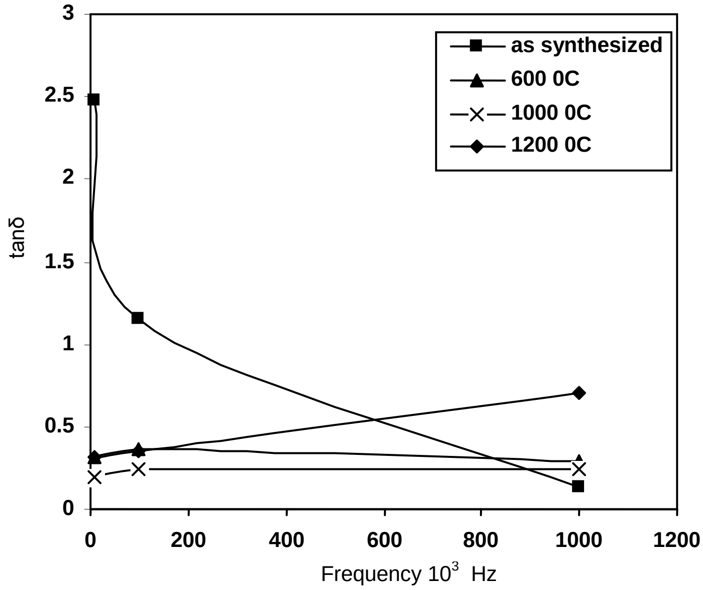


Fig.7 Loss factor versus frequency for various annealing temperatures.

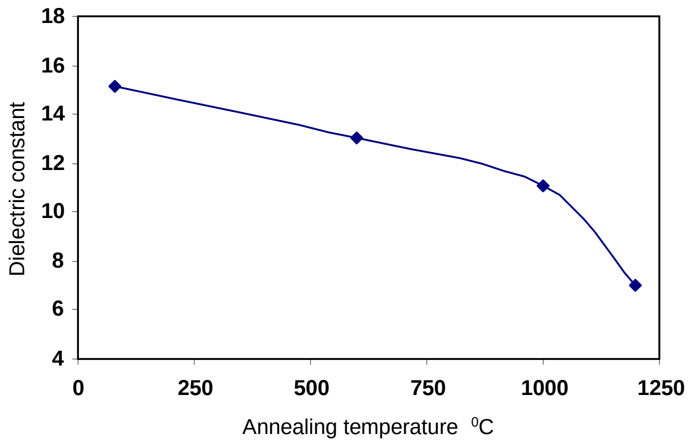


Fig.8 Dielectric constant versus annealing temperature at 10 KHz at room temperature.

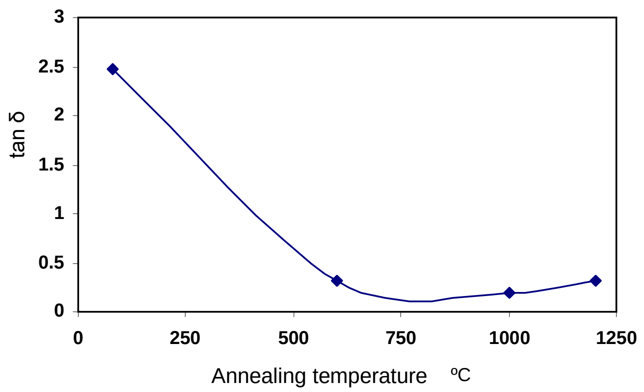


Fig.9 Loss factor versus annealing temperature at 10 KHz

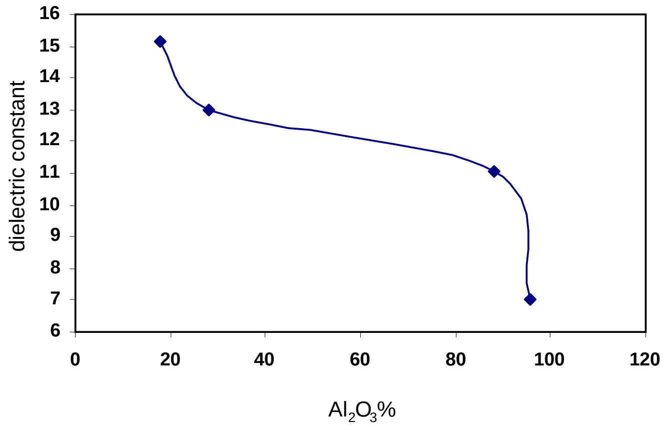


Fig.10 Dielectric constant versus Al_2O_3 % at room temperature