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Study on Variation of Ferroelectric Parameters of Dielectric Materials $ZrTiO_4$ +Al with Quality Factor and Resonant Frequency

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ABSTRACT

The variation of dielectric constant with temperature of mixed compound of Zirconium titanate with aluminium and other dielectric parameters have been measured between the temperatures 35°C to 300°C using the capacitance bridge model ZENITH-M92A and Q meter at the frequency of 2000 Hz in which percentage of Al is from 20 to 35. The solid solution of mixed compound of Zirconium titanate with aluminium has excellent dielectric property such as dielectric constant of nearly $\varepsilon = 7000$, quality factor Q_f = 25000GHz and temperature coefficient of resonant frequency τ_f = 75 ppm/°C which is very much useful in ceramics engineering and communication system. In the measurement, it have been observed that the compound has lower value of dielectric constant ($\varepsilon = 1580$) below 36ºC, which rises upto a value of 5500 at the moderate temperature of 76ºC. After this temperature the dielectric constant of compound decreases upto the value of nearly 1500 at the temperature of 88ºC and a high peak is obtained at the temperature of 150°C ($\varepsilon = 10000$) in the heating cycle curve with some fluctuations. When the variation of dielectric constant was studied in cooling cycle the peak was observed at 140ºC($\varepsilon = 10000$), above and below this temperature, dielectric constant decreases with some intermediate fluctuations. The cooling cycle curve does not follow heating curve because of the temperature relaxation of the compound. Annealing Zirconium titanate with aluminium increases the order parameter and improves the dielectric quality factor. Many investigation showed as the mixed compound as a useful temperature-stable dielectric ceramic device and by increasing aluminium content in the mother compound, its quality factor also increases with a little effect on dielectric constant. One of the advantages of $ZrTiO_4$ with aluminium is that by varying the aluminium content, we can control temperature coefficient of resonant frequency τ_f without drastically affecting the other properties.

Introduction

The recent progress in microwave telecommunication, satellite broadcasting and intelligent transport systems (ITS) has resulted in an increasing demand for dielectric resonators (DRs), which are low loss ceramic pucks used mainly in wireless communication devices[1]. With the recent revolution in mobile phone and satellite communication systems using microwaves as the carrier, the research and development in the field of device miniaturization has been one of the biggest challenges in contemporary Materials Science. This revolution is apparent on a daily basis in the ever increasing number of cell phone users. The recent advances in materials development has led to these revolutionary changes in wireless communication technology. Dielectric oxide ceramics have revolutionized the microwave wireless communication industry by reducing the size and cost of filter, oscillator and antenna components in applications ranging from cellular phones to global positioning systems. Wireless communication technology demands materials which have their own specialized requirements and functions. The importance of miniaturization cannot be overemphasized in any hand-held communication application and can be seen in the dramatic decrease in the size and weight of devices such as cell phones in recent years. This constant need for miniaturization provides a continuing driving force for the discovery and development of increasingly sophisticated materials to perform the same or improved function with decreased size and weight [2].

A DR is an electromagnetic component that exhibits resonance with useful properties for a narrow range of frequencies. Dielectric Resonators (DR) are dielectric bodies of high permittivity and high Q-factor that can be used as energy storage devices. Ceramic DRs are usually prepared in the form of cylindrical or rectangular pucks by the sintering process. They are much smaller in size compared to its metallic counterpart. The three important characteristics of an ideal DR are high relative permittivity or dielectric constant (ε_r) for resonator applications and low ε_r for millimeter wave applications, low dielectric loss tan δ and low coefficient of temperature variation of the resonant frequency τ_f[3].

The term ‘‘quality factor’’ is more commonly associated with microwave resonators. Quality factor, or Q, is a measure of the power loss of a microwave system. The name quality

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factor is used for the reciprocal of the tan δ. One should carefully distinguish this quantity from the Q-factor of a resonator which is defined as

Q = 2 π[(maximum energy stored per cycle)/(Average energy dissipated per cycle)]

The temperature coefficient of resonant frequency τ_f is the parameter which indicates the thermal stability of the resonator. The τ_f indicates how much the resonant frequency drifts with changing temperature. The electronic device with microwave resonators requires τ_f values as close to zero as possible. Microwave circuits will normally have some low characteristics τ_f, so the resonator components which go into them are required to compensate for the inherent drift. For this reason, the τ_f values of resonators required are typically non-zero but with some low finite value. The origin of τ_f is related to linear expansion coefficient α_L which affects the resonator dimensions and its dielectric constant variation with temperature. Mathematically

$$\tau_f = - \alpha_L - \tau_{E/2}$$

where τ_E is the temperature coefficient of the permittivity and α_L is the linear thermal expansion coefficient of the dielectric material which is usually positive[4].

Experimental

The compound has been procured from Sigma-Aldrich (India), Bangalore. The chemical was grinded into the fine powder in an agate mortar, avoiding direct sunlight and preferably the most of the sample preparation was done at night. The pellets were prepared with compression machine (Flextural Testing Machine CAT No.AIM-313, S.No.91070 AIMIL Associated, India), having pressure range 0-10 tonne wt/cm^2. A suitable die was used having rectangular Cross-Sectional area of the piston =2.50cm^2.The polishing of the pellets has been done to obtain smooth parallel surface to be used for electrode formation polishing of the crystal introduces electrical charges inside the material. These charges and strains are to be removed, which we did by the process of annealing of the sample. In this process the pellets were kept in a suitable furnace at nearly 2/3 of their melting points for sufficient times (generally 8-10 hours). The most of the irreproducibility was removed by annealing and therefore this process was necessarily done. The electrodes were formed using colloidal silver paints [5, 6].

The sample holder loaded with pellet is kept into the furnace such that it lies very near to the middle part of the furnace. A good quality thermometer, precisely calibrated is used to record the temperature. This thermometer is adjusted with the help of stand in such a way that it touches the metallic part of sample holder to record the exact temperature of sample. The usual substitution method i.e. with and without the specimen in suitable sample holder is used. The sample holder was directly fastened to the capacity measuring unit (ZENITH-M92A) and Q meter. There are some process from which we have done the measurement of the quality factors of dielectrics.. Wang [7] prepared $ZrTiO_4$ ceramics with additives such as ZnO, CuO and Y_2O_3 and reported that the microstructure and microwave dielectric properties are sensitive to the presence of additives and processing conditions. Single phase $ZrTiO_4$ was formed on annealing the milled powders and the grain size increased with prolonged annealing [8].

Result and Discussion

The variation of dielectric constant with temperature of Zirconium titanate with aluminium and other dielectric parameters have been measured between the temperatures 35°C to 300°C using the capacitance bridge model ZENITH-M92A and Q meter at the frequency of 2000 Hz in which percentage of Al is from 20 to 35. The solid solution of mixed compound of Zirconium titanate with aluminium has excellent dielectric property such as dielectric constant of nearly ε = 7000, quality factor Q_f = 25000GHz and temperature coefficient of resonant frequency τ_f = 75 ppm/°C which is very much useful in ceramics engineering and communication system. In the measurement, it have been observed that the compound has lower value of dielectric constant (ε = 1580) below 36ºC, which rises upto a value of 5500 at the moderate temperature of 76ºC. After this temperature the dielectric constant of compound decreases upto the value of nearly 1500 at the temperature of 88ºC and a high peak is obtained at the temperature of 150°C (ε = 10000) in the heating cycle curve(Fig.1) with some fluctuations. When the variation of dielectric constant was studied in cooling cycle the peak was observed at 140ºC(ε = 10000), above and below this temperature, dielectric constant decreases with some intermediate fluctuations. The cooling cycle curve does not follow heating curve because of the temperature relaxation of the compound. The curve for the cooing cycle is shown in the Fig.2[9]. Mixed compound of Zirconium titanate with aluminium has many applications usually in wireless communications technology and also in high temperature pigments in chemical industry. The $ZrTiO_4$ is orthorhombic with lattice parameters

a=4.806 Å, b=5.447 Å, c=5.032 Å

and two formula units in the cell with theoretical density of 5.15 gm/cm^3[10].

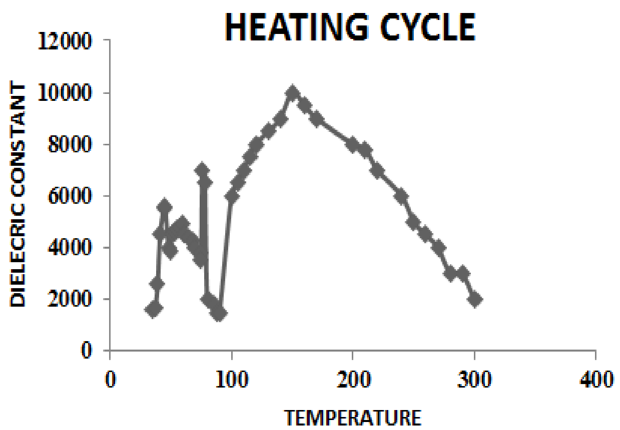

Fig 1. Temperature variation of dielectric constant of mixed compound of zirconium titanate with aluminium.

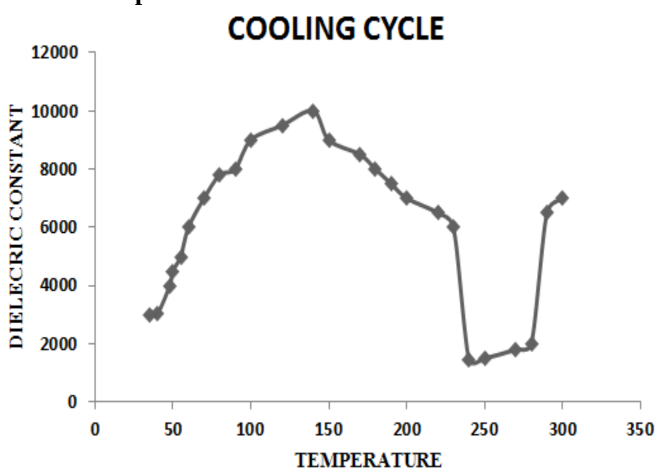

Fig 2. Temperature variation of dielectric constant of mixed compound of zirconium titanate with aluminium.

Fig. 3 shows the variation of τ_f as a function of Aluminium content Zirconium titanate . By increasing Aluminium content in the mother compound, its quality factor also increases with bump with a little effect on dielectric constant. One of the advantages of $ZrTiO_4$ with aluminium is that by varying the aluminium content, we can control temperature coefficient of resonant frequency τ_f without drastically affecting the other properties [11].

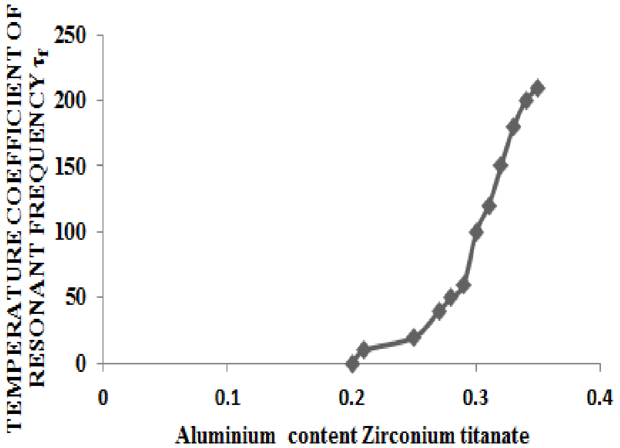

FIG 3. Variation of τ_f as a function of Aluminium content Zirconium titanate.

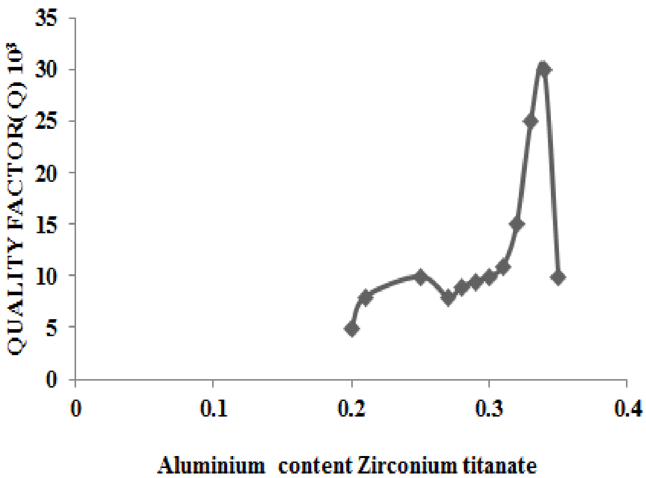

Fig 4. Variation Of Quality Factor With Aluminium Content Zirconium Titanate.

Fig.4 shows the variation of quality factor as a function of Aluminium content Zirconium Titanate. Although the Q_f appears to increase with larger grains produced by longer sintering times, it is not the grain size itself that is controlling the Q_f. The effect of annealing the $ZrTiO_4$ ceramics on the dielectric properties was found that slow-cooled ceramic has a much higher quality factor as compared to rapidly cooled ceramic[12]. Rapid cooling from the sintering temperature yielded a disordered structure having a low Q_f value. Zirconium titanate with aluminium is an important dielectric material with excellent properties useful for applications in communication technology[13].

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