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Non Linear Dielectric Phenomenon in $BaTiO_3$ - Bi $(Ti_{1-x}Mg_x)O_3$. Ceramic Material

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ABSTRACT— ***The distinguishing characteristic of an insulator is its resistivity. An insulator has a higher resistivity than a semiconductor or conductor and has high dielectric properties. The adjustable dielectric property is characterized by a change in the permittivity of the dielectric under an applied electric field, this phenomenon is called "dielectric nonlinearity" (dielectric tunability properties). With these properties, a new phenomenon will arise adjustable dielectric properties in dielectric ceramic materials. The aim of this research is to observe the non-linear dielectric phenomenon in*** $BaTiO_3$-$Bi(Ti_{1-x}Mg_x)O_3$ ***ceramic material with x= 0.5, 0.6, 0.7, 0.8 and 0.9. Ceramic materials were synthesized using the sol-gel method which produced nano-sized powders. Impedance data was measured using an impedance analyzer (Solartron SI 1260, Solartron Metrology, West Sussex, UK) in a frequency range of 100 Hz to 500 Hz with an ac measuring voltage of 0.1 V. Dielectric tunability properties were measured from 0ºC to 250ºC using an automatic component analyzer. at 10 kHz. All samples show the phenomenon of dielectric tunability with different maximum permittivity. The highest permittivity was obtained at x= 0.6 but the highest transition temperature where non-linear phenomena occurred was found in the sample with x= 0.5.***

Keywords— non-linear dielectric, tunability, permittivity

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1. INTRODUCTION

Research on advanced ceramic materials continues to grow and what has been consistently researched in the last 5 years is insulating materials that are widely applied to electronic devices. The distinguishing characteristic of an insulator is its resistivity. An insulator has a higher resistivity than a semiconductor or conductor and has high dielectric properties [1,2]. The adjustable dielectric property is characterized by a change in the permittivity of the dielectric under an applied electric field, this phenomenon is called "dielectric nonlinearity" (dielectric tunability properties). With these properties, a new phenomenon will arise adjustable dielectric properties in dielectric ceramic materials. Several researches have been developed to improve dielectric tunability properties, including: $Ba_{1-x}Sr_xTiO_3$(BST), $BaTi_{1-x}Zr_xO_3$ (BZT), $Pb_{1-x}Sr_xTiO_3$ (PST). Bi $(Ti_{1-x}Mg_x)O_3$, which is the result of engineering existing materials with the aim of improving dielectric tunability properties by referring to the transition temperature where the permittivity value changes. The nano size of the powder is needed to increase the surface area which can increase the interaction of ferroelectric atoms [3]. The technology used for this synthesis process is sol-gel which is an easy technology, requires simple equipment and a low process temperature needed and can produce nano-sized powders.

With this research, it is hoped that a solution can be obtained to obtain high dielectric properties in a new ceramic material based on $BaTiO_3$ - Bi $(Ti_{1-x}Mg_x)O_3$. The application of materials with this property is as a separator of electrical conductors so that electricity cannot pass between conductors, in this case as a ceramic capacitor separator for the main electrical circuit board of a computer device or in a television liquid crystal display.

Recently electrically adjustable dielectrics have attracted much interest for their potential applications as capacitors, phase shifters, adjustable filters and voltage controlled oscillators [4-6]. The adjustable dielectric property is characterized by a change in the permittivity of the dielectric under an applied electric field, and this phenomenon is called "dielectric nonlinearity". The permittivity of the dielectric usually decreases with increasing the applied electric field. However, certain ferroelectric compositions of $PbSrZrTiO_3$ (for example) exhibit abnormal additional permittivity changes when placed in an applied electric field [7]. Some researchers regard this abnormal behavior as electric field-induced antiferroelectric to ferroelectric phase transition [8] due to the anisotropic nature of ferroelectricity [9]. Drougard [10] considered the increase in dielectric permittivity in the $BaTiO_3$ single crystal to be due to the loss of the 180° domain wall under the refractive field. Therefore, the origin of this abnormal nonlinear dielectric behavior is still controversial today and still needs to be investigated further.

The $BaTiO_3$ - $Bi(Ti_{1-x}Mg_x)O_3$ system, (BT-BMT) has a pseudocubic perovskite structure and temperature dependence like a relaxor of permittivity [11,12]. Furthermore, Wada [13] found that the ceramics of the BT-BMT system exhibit relaxant-like characteristics with a maximum dielectric temperature of as high as 360°C for 0.5BT–0.5BMT ceramics and a piezoelectric constant of 60 pC/N for 0.4BT-0.6BMT. Randall [14] built a prototype multilayer capacitor with dimensions of 18 mm × 317 mm × 34 mm using BT-BMT and its capacitance reaches 12.5 nF at 1 kHz. Therefore, in this study, the $BaTiO_3$ - $Bi(Ti_{1-x}Mg_x)O_3$, (BT-BMT) dielectric system was investigated and characterized to determine the occurrence of non-linear dielectric phenomena.

2. METHOD AND MATERIALS

BT-BMT ceramics were made using the sol gel method using the basic ingredients of $Bi(NO_3)_2$ (99.99%), TiO_2 (99.9%), MgO (99.9%) and $BaCO_3$ (99.9%) and weighed to prepare proper molar ratio x=0.5, 0.6, 0.7, 0.8 and 0.9. The powder mixture was mixed with aquabidestilate and then heated at 80-90°C until a gel was formed. The gel was then calcined at 950°C for 4 hours to remove H_2O and other gases. After the calcination process, a powder is obtained which must be ground before being formed into pellets. The pellets were then sintered at a temperature of 1200°C for 5 hours. The sintered pellets were then polished to a final thickness of 0.5 mm for dielectric characterization. Silver electrodes of suitable configuration were coated on both polished surfaces and fired at 550°C for 30 minutes to form a metal-insulating-metal (MIM) capacitor for electrical testing. Weak-field dielectric response at a signal level of 500 mV/mm was measured using a precision impedance analyzer (4294A, Agilent, Santa Clara, CA, USA) associated with a temperature controller (TP94, Linkam, Surrey, UK) over a temperature range of 0 – 250°C at a heating rate of 2°C/min. The impedance data was then measured using an impedance analyzer (Solartron SI 1260, Solartron Metrology, West Sussex, UK) in a frequency range of 100 Hz to 500 Hz with an AC measuring voltage of 0.1 V. Dielectric tunability properties were measured at a temperature of 0 – 250°C using an automatic component analyzer. (TH2818, Tonghui, Changzhou, China) at 10 kHz. A blocking circuit is adopted to protect the analyzer from the applied bias voltage.

3. RESULTS AND DISCUSSION

Non-linear dielectric phenomena were found in all samples (x= 0.5, 0.6, 0.7, 0.8 and 0.9) as shown in Figure 1-4. The permittivity of the dielectric first increases until it reaches the threshold of the electric field, and then suddenly decreases. The abnormal nonlinear dielectric behavior is caused by the nanopoles being clamped by the oxygen vacancies in the sample and cannot be easily reoriented under an applied electric field, besides that the Bi atoms tend to form an insulating phase at the grain boundaries which weakens the effective electric field.

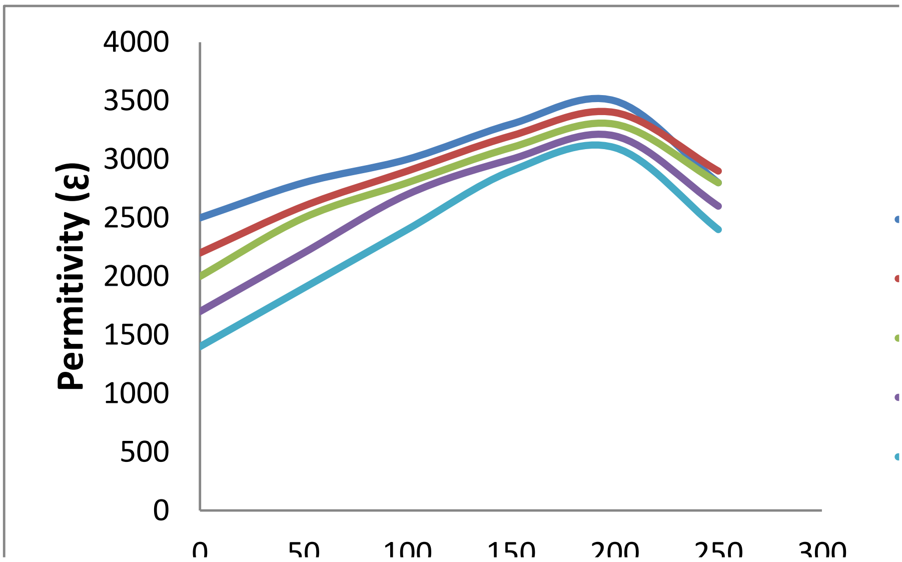

Figure 1. Phenomenon of Non Linear Dielectric for BT-BMT, x = 0.5

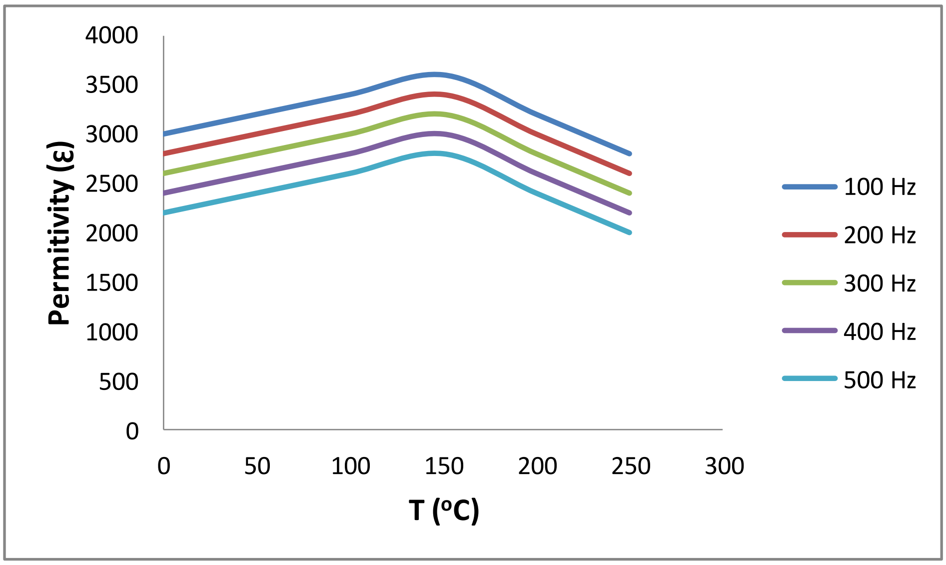

Figure 2. Phenomenon of Non Linear Dielectric for BT-BMT, x = 0.6

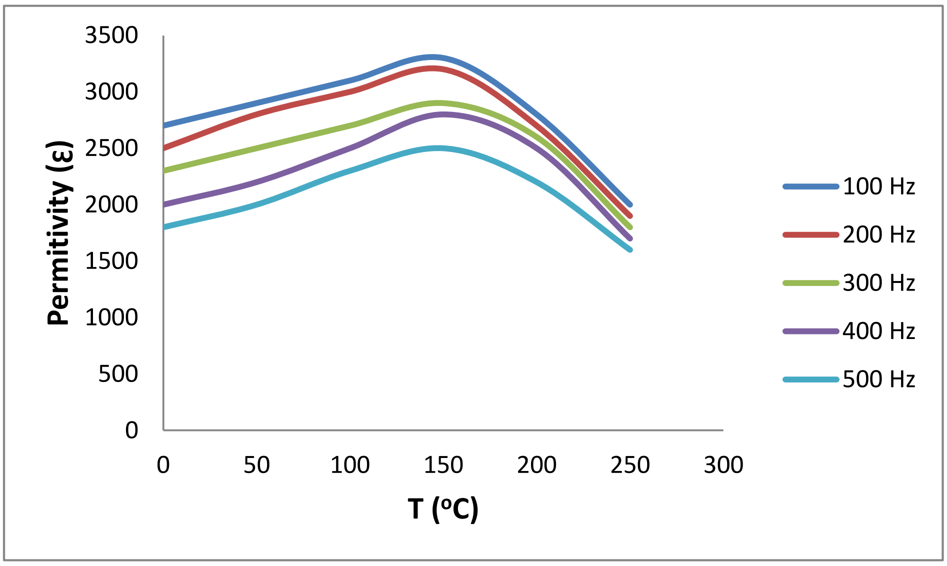

Figure 3. Phenomenon of Non Linear Dielectric for BT-BMT, x = 0.7

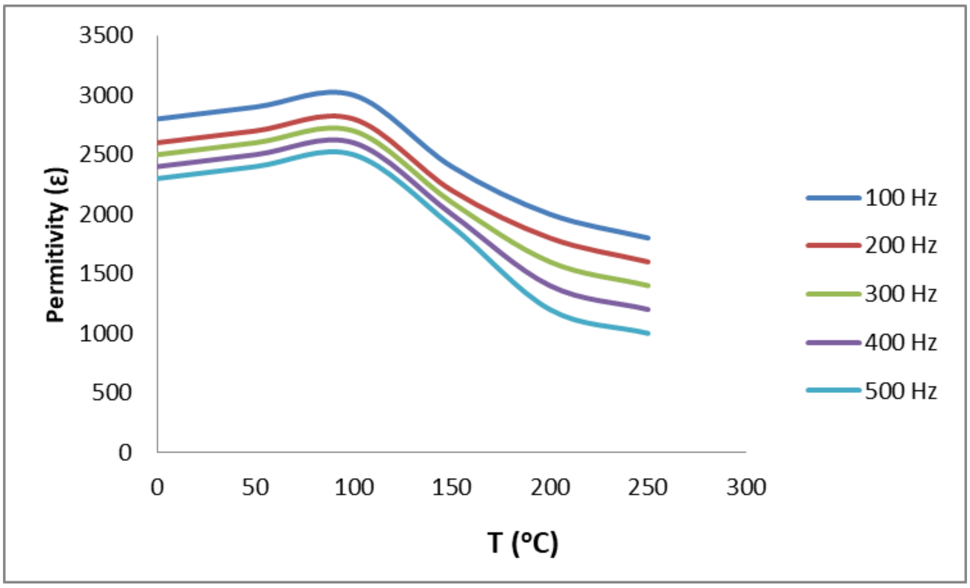

Figure 4. Phenomenon of Non Linear Dielectric for BT-BMT, x = 0.8

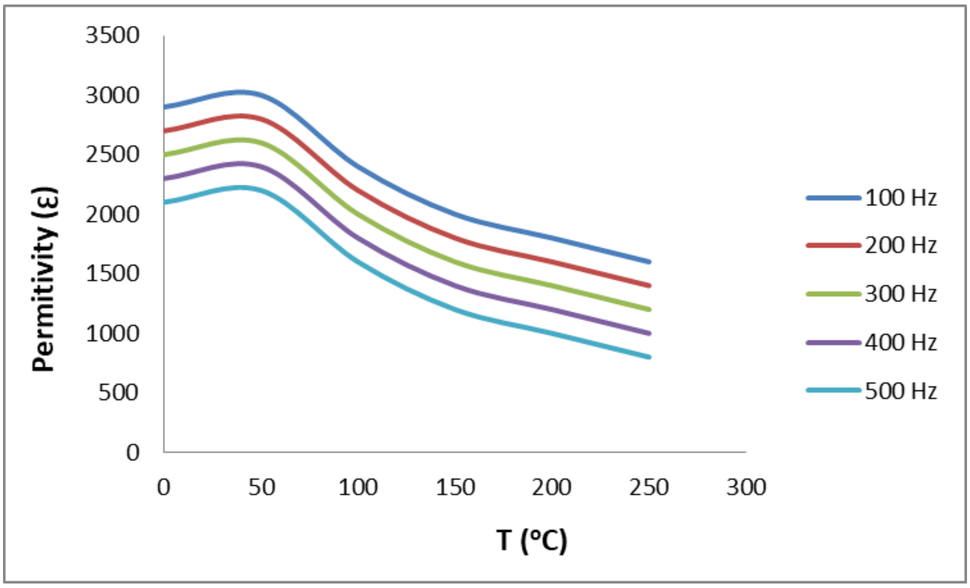

Figure 5. Phenomenon of Non Linear Dielectric for BT-BMT, x = 0.9

Figure 4 shows the temperature dependence of the dielectric permittivity (ε) for all BT-BMT samples measured at various frequencies (100 – 500 Hz) at a temperature of 0 - 250ºC. A strong dielectric dispersion or frequency dependence of the dielectric properties was observed, indicating that there was a difference in transition temperature. In BT-BMT, x= 0.5 the decrease in permittivity value occurs at the highest temperature at the transition temperature of 200ºC, especially at lower frequencies. While the highest dielectric permittivity value is owned by the BT-BMT sample, x = 0.6. The formation of a dielectric permittivity versus temperature curve, confirms that the sample contains a mixture of tetragonal and pseudocubic structures [15]. The peak at lower temperatures corresponds to the phase transition from pseudocubic to cubic, while the peak at higher temperatures corresponds to the phase transition from tetragonal to cubic. The frequency plateau represents the bulk permittivity of the sample; with decreasing frequency, the curve tends to be steeper which is attributed to the grain boundary contribution. Randall [14] investigated the dielectric versus temperature spectrum (1-x) of BMT-x BT and suggested that the tetragonal phase and pseudocubic phase might coexist at x = 0.1 and change to a pseudocubic phase at x above 0.20.

4. CONCLUSION

$BaTiO_3$ - Bi $(Ti_{1-x}Mg_x)O_3$ (BT-BMT), x= 0.5, 0.6, 0.7, 0.8 and 0.9 ceramics were made using the sol gel method and showed a non-linear dielectric phenomenon as indicated by the appearance of an electric permittivity versus temperature curve. This curve appears due to the nanopoles being clamped by the oxygen vacancies in the sample and cannot be easily reoriented. All dielectric permittivity curves shift and the largest shift (right direction) occurs at x= 0.5 at a temperature of 200°C, while the highest permittivity value occurs at x= 0.6. Therefore, it is recommended that the use of this material for electronic devices is at x = 0.5 – 0.6.

5. ACKNOWLEDGEMENT

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