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Comparison of Ferroelectric phase transition in $BaSr_4RTi_3V_7O_{30}$ (R= Gd, La) ceramics

B. B. MOHANTY, [1] P. S. SAHOO, [1†] R. N. P. CHOUDHARY[2]
[1] Department of Physics, Betnoti College Betnoti, Mayurbhanj, Odisha, India
[2] Department of Physics ITER, Bhubaneswar, Odisha, India
[†]Corresponding Author: email: ps_rilly@yahoo.com

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Abstract : Looking to the demand of materials having high dielectric constant and low loss, are very much useful for their effective use as transducers, actuators, capacitors and also in memory devices, polycrystalline samples of $BaSr_4RTi_3V_7O_{30}$ (R = Gd, La) are synthesized by a high temperature solid state reaction technique. Formation of single-phase orthorhombic structures at room temperature having average crystallite size of the order of some nanometer for all the compounds are confirmed from preliminary XRD analyses of these compounds. Detailed dielectric study in a wide temperature range (33–500^{o}C) shows ferro to para phase transition for Gd doped compound and no such transition is observed for La doped compound.

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1. Introduction

The discovery of ferroelectricity in $BaTiO_3$ boosted the researchers to study a large number of oxides of different structures such as perovskite, tungsten bronze, layer structure etc, for their applications in some devices like transducers, actuators, multi-layer capacitors ferroelectric random access memory and display etc. In this context some ferroelectric oxides of tungsten bronze (TB) structural family have widely been investigated because of their favorable dielectric [1], electro-optic [2], pyroelectric [3], piezoelectric [4], nonlinear optic [5], acoustic optic [6] properties. The TB structure consists of a framework of distorted BO_6 octahedral sharing corners in such a way that three different types of interstices (A, B and C) are available for a wide variety of cations occupying in a general formula $(A_1)_2(A_2)_4(C)_4(B_1)_2(B_2)_8O_{30}$. It has been found that different ionic substitutions at the above-mentioned sites can play an important role to tailor their physical properties. In this view, we have synthesized and studied the dielectric and conductivity properties of $BaSr_4GdTi_3V_7O_{30}$ and $BaSr_4LaTi_3V_7O_{30}$ ceramics.

2. Experimental details

The polycrystalline sample $BaSr_4GdTi_3V_7O_{30}$ **(**BSGTV**)** and $BaSr_4LaTi_3V_7O_{30}$ (BSLTV) were prepared using high purity (>99.9%) precursors; $BaCo_3$, $SrCo_3$, Gd_2O_3, La_2O_3, TiO_2, V_2O_5 (M/S Sarabhai M. Chemicals, India), by a high-temperature solid-state reaction technique. These ingredients (taken in suitable stoichiometry), were thoroughly mixed and grounded in dry and wet (methanol) medium for 3 h each in an agate mortar. The mixed powders were calcined at optimized temperature and time (950 °C for 24 h) in air. The calcined powder was mixed with PVA (poly vinyl alcohol) binder, grounded and subsequently pressed into pellets (about 10 mm in diameter and 1-2 mm thickness) under uniaxial pressure of 5×10^6 N/m^2. The desired ceramic materials were obtained by sintering the pellets in air at 1000°C for 12 h. The sintered pellets were then polished on both the faces flat and parallel. The pellets were then electroded with high purity air-drying silver paste and then dried at 150 ^{0}C for 4 h, before taking any electrical measurements. Room temperature X-ray diffraction (XRD) data (pattern) of the material obtained in a wide range of Bragg angle 2θ ($20^0\leq 2\theta \leq 80^0$), at a scanning speed of 3^0 min^{-1} by an X-ray diffractometer (Rigaku, Miniflex) with CuKα radiation (λ= 1.5405Å) at room temperature confirms the formation of desired compounds. Detailed of Structural properties of both the compounds are reported earlier [7, 8]. The dielectric studies were carried out in the temperature range of 30^0 -500^0C and wide frequency range of 100Hz to 1MHz, using a computer-controlled impedance analyzer(PSM 1735, model: N 4L).

3. Results and discussion

3.1. Dielectric properties

Temperature dependence of relative dielectric constant (ε_r) and dielectric loss of BSGTV and BSLTV is as shown in fig. 1. It is evident from the figure that $BaSr_4GdTi_3V_7O_{30}$ compound shows two frequency independent dielectric anomalies (at 185^{o}C and 428^{o}C). The first peak indicates the occurrence of ferroelectric–paraelectric phase transition. The second peak may be due to ionic transition [9, 10]. But no such anomaly is observed for $BaSr_4LaTi_3V_7O_{30}$ compound in the experimental range. In this compound there is a very small change in the values of ε_r up to 300 ^{0}C then increases rapidly with increasing temperature. It is also observed that Gd modified compound have high dielectric constant and loss as compared to La modified compound. From the tan δ versus temperature plot (Fig. 1 (insert)), similar nature of variation was observed. The loss is very small below $T_{c,}$ for BSGTV compound and a change in slope observed at higher temperature.

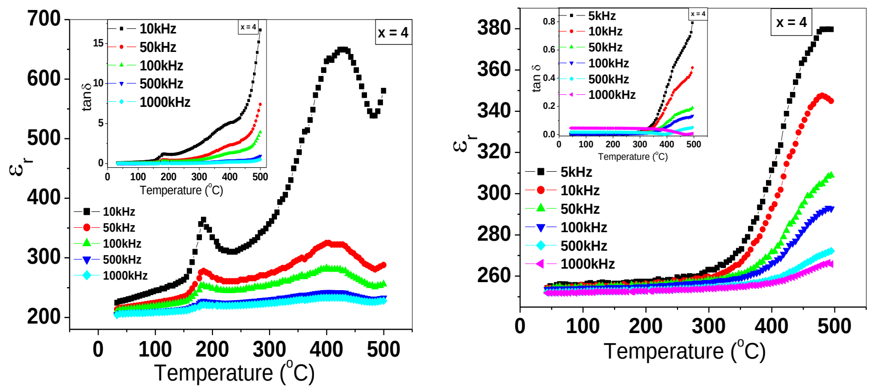

Fig.1. Variation of ε_r and tanδ with temperature of $BaSr_4GdTi_3V_7O_{30}$ (left) and $BaSr_4LaTi_3V_7O_{30}$ (right) compounds at some selected frequencies.

3.2. Hysteresis study

Fig.2. Shows the room temperature variation of polarization as a function of electric field on poled sample of $BaSr_4RTi_3V_7O_{30}$ (R = Gd, La) compounds. The loop area of BSGTV ceramics is very small indicated by very small remanent polarization (2Pr = 0.012 μC/cm^2 at an applied electric field of and 5.6 kV/cm) in it. But a proper hysteresis loop is not obtained because of the lossy characteristics of the material. For La modified compounds no such loop is observed. This is in good agreement with dielectric analysis, as no phase transitions are observed for this compound.

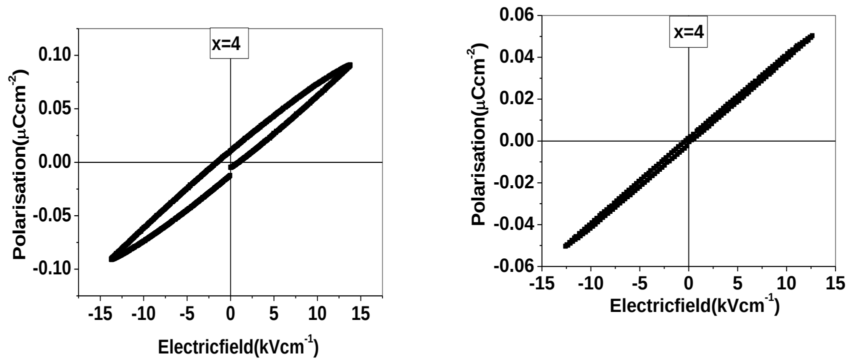

Fig. 2. P~E loop of of $BaSr_4GdTi_3V_7O_{30}$ (left) and $BaSr_4LaTi_3V_7O_{30}$ (right) compounds at room temperature

4. Conclusion

The BSGTV and BSLTV of tungsten–bronze family are prepared, and their dielectric and polarization characteristics are investigated. Detailed dielectric study shows two frequency independent dielectric anomalies (at 185^{o}C and 428^{o}C) for Gd doped compound, but no such transition is observed for La doped compound. Gd modified compound have high dielectric constant and loss as compared to La modified compound. For BSGTV hysteresis loop is obtained with small remanent polarization, but no such loop is observed for La modified compound.

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