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Investigation on dielectric properties of Al_2O_3 , ZnO and MgO doped $Ba_{0.7}Sr_{0.3}TiO₃$ ceramics

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ABSTRACT

Being an insulator material with high relative permittivity, Barium Strontium Titanate (BST) ceramics can be used extensively in many applications if its dissipation factor is reduced. In this paper, we investigate dielectric properties of $Ba_{0.7}Sr_{0.3}TiO₃$ ceramics when doped with Al₂O₃, ZnO and MgO. At first, $Ba_{0.7}Sr_{0.3}TiO₃$ ceramics was synthesized using conventional solid-state reaction technique. Further the $Ba_{0.7}Sr_{0.3}TiO₃$ ceramic samples were doped with 0.5, 1.0, 2.0, 4.0 and 8.0 wt% Al₂O₃. Similarly $Ba_{0.7}Sr_{0.3}TiO₃$ ceramic samples were doped with same weight percentages of ZnO and then MgO. After making pellets of all these samples, silver paste was coated for top and bottom electrodes to form ceramic based Metal-Insulator-Metal (MIM) capacitors. The dielectric properties were measured at room temperature using Hioki 3532-50 LCR Hi-Tester. It is noted that relative permittivity increases with dissipation factor (dielectric loss) irrespective of doping material and doping concentration. The dissipation factor achieved by 2.0 wt% ZnO doped $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics is 0.0087, which is promisingly very low. Further it is observed that 2.0 wt% ZnO doped $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics possesses highest figure of merit (relative permittivity to dissipation factor ratio) and hence optimum performance among the doping materials and doping concentrations considered. 2020 Elsevier Ltd. All rights reserved.

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1. Introduction

The insulator materials with high relative permittivity help in reducing the size of electronic devices due to smaller capacitive components $[1]$. Owing to high relative permittivity, BaSrTiO₃ ceramics are good choice for many tunable applications. Even then dissipation factor, the important parameter of BaSrTiO₃ ceramics in many dielectric applications, is much higher than 0.02 which restrict their extensive usage $[2,3]$. The dissipation factor should be lower than 0.01 to use in RF and microwave applications $[4,5]$, microwave substrate application [\[6\],](#page-4-0) phase shifter application [\[7\]](#page-4-0), energy storage application [\[8\]](#page-4-0) and power application [\[9\].](#page-4-0) Hence it is very crucial to reduce the dissipation factor of BaSrTiO₃ ceramics materials [\[10\].](#page-4-0) Studies reveal that small concentrations of

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dopants result in drastic changes in the properties of BaSrTiO₃ ceramics $[11]$. BaSrTiO₃ ceramics have been doped with some materials such as Co [\[12\],](#page-4-0) Ti [\[13\]](#page-4-0), La [\[14\]](#page-4-0), W [\[15\]](#page-4-0), Al [\[16\]](#page-4-0), Ce [\[17\]](#page-4-0), Cr [\[18\]](#page-4-0), Bi [\[19\]](#page-4-0), Mg [\[20\]](#page-4-0), Zr [\[21\],](#page-4-0) Mn [\[22\]](#page-4-0) and Fe [\[23\]](#page-4-0) to lower the dissipation factor.

In this work, dielectric properties of $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics when doped with Al_2O_3 , ZnO and MgO are investigated. The dopant solubility in $Ba_{0.7}Sr_{0.3}TiO₃$ ceramics depends mainly on its ionic radius [\[24\]](#page-4-0). The ionic radii of Ba²⁺, Sr²⁺ and Ti⁴⁺ are 1.36 A^o, 1.16 A^o and 0.68 A^o respectively [\[25,26\]](#page-4-0). The ionic radii of Al^{3+} , Zn²⁺ and Mg^{2+} ions are 0.675 A°, 0.88 A° and 0.86 A° respectively [\[27\]](#page-4-0) which are comparable to that of Ti⁴⁺. This enhances the chances for Al^{3+} , Zn^{2+} and Mg^{2+} ions to replace Ti⁴⁺ ions from BST perovskite structure during the process of doping.

The melting points of Al₂O₃, ZnO and MgO are 2072 °C, 1975 °C and 2852 \degree C respectively which are well below the sintering temperature of BST samples (1275 \degree C) and hence the synthesis of ceramics won't be affected adversely. The changes in relative

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permittivity and dissipation factor with doping concentrations are also investigated.

2. Experimental procedure

BaCO₃ (Spectrum, 99%, 197.35 g/mol), SrCO₃ (Spectrum, 99%, 147.63 g/mol) and TiO₂ (Spectrum, 98%, 79.886 g/mol) nanopowders; all of ACS reagent grade were used as precursors for $Ba_{0.7}Sr_{0.3}TiO₃$ nanopowder synthesis. Stoichiometric proportions of BaCO₃ and TiO₂ nanopowders were blended for 1 h in agate mortar. Similarly stoichiometric proportions of $S_{rcO₃}$ and TiO₂ nanopowders were blended for 1 h. Both mixtures were calcined for 4 h at 1100° C in muffle furnace. The melts were blended for 1 h to get BaTiO₃ and SrTiO₃ nanopowders. Stoichiometric proportions of BaTiO₃ and SrTiO₃ nanopowders were blended for 1 h and then calcined for 4 h at 1100 \degree C to get Ba_{0.7}Sr_{0.3}TiO₃ nanopowder. The stoichiometric proportions of BaCO₃ and TiO₂ nanopowders required for synthesizing 1 g BaTiO₃ nanopowder are 0.8462 g and 0.3425 g respectively. The stoichiometric proportions of $SrCO₃$ and TiO₂ nanopowders required for synthesizing 1 g SrTiO₃ nanopowder are 0.8045 g and 0.4353 g respectively. The stoichiometric proportions of BaTiO₃ and SrTiO₃ nanopowders required for synthesizing 1 g $Ba_{0.7}Sr_{0.3}TiO_3$ nanopowder are 0.7478 g and 0.2522 g respectively.

The synthesized $Ba_{0.7}Sr_{0.3}TiO_3$ sample was blended with 0.5, 1.0, 2.0, 4.0 and 8.0 wt% Al_2O_3 (Nice, 99%, 101.96 g/mol). Further $Ba_{0.7}Sr_{0.3}TiO₃$ sample was blended with 0.5, 1.0, 2.0, 4.0 and 8.0 wt% ZnO (Merck, 99%, 81.37 g/mol) and then with 0.5, 1.0, 2.0, 4.0 and 8.0 wt% MgO (Nice, 99%, 40.30 g/mol). After one hour of blending, these samples were calcined for 4 h at 1100C. The calcined samples were blended for 1 h and were die-pressed into pellets after mixing each sample with the binder (4 wt% aqueous PVA) using Carver auto series automatic hydraulic laboratory press with digital control system. The pellets were then sintered for 2 h at 1275 \degree C in Nabertherm sintering furnace.

Further silver (Ag) paste was painted over top and bottom surfaces of pellets to make Metal-Insulator-Metal capacitor. Copper wires at top and bottom electrodes were fixed for capacitor leads. The relative permittivity and dissipation factor were measured at room temperature using Hioki 3532-50 LCR Hi-Tester.

3. Results and discussions

The relative permittivity and dissipation factor of intrinsic and doped $Ba_{0.7}Sr_{0.3}TiO₃$ ceramics were measured at room

temperature. Fig. 1 compares relative permittivity and dissipation factor of intrinsic and Al_2O_3 -doped $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics with different doping concentrations measured at 10 kHz. From Fig. 1, it is observed that the relative permittivity and dissipation factor of $Ba_{0.7}Sr_{0.3}TiO₃$ ceramics decreased drastically when doped with 0.5 wt% Al_2O_3 . When doped with 1.0 wt% Al_2O_3 , relative permittivity increased and dissipation factor decreased. This may be due to increase in oxygen vacancies. The lowest dissipation factor is observed when doped with 2.0 wt% Al_2O_3 . At this doping concentration, the surplus active electrons are exactly compensated [\[16\]](#page-4-0). The threshold of 1.0 wt% for relative permittivity and 2.0 wt % for dissipation factor observed in this work is similar to that achieved by Wu et al. [\[28\]](#page-4-0) at same frequency and temperature.

The relative permittivity and dissipation factor of intrinsic and ZnO-doped $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics with different doping concentrations measured at 10 kHz are given in Fig. 2. It is noted that relative permittivity increases till 2.0 wt% ZnO and decreases thereafter. This is due to smaller grains and higher density at low ZnO doping concentrations. When doping concentration increases grains will be higher and density will be lower. It is observed from Fig. 2, the relative permittivity and dissipation factor of $Ba_{0.7}Sr_{0.3}$ $TiO₃$ ceramics dramatically decreased when doped with 0.5 wt% ZnO as in the case of dopant material Al_2O_3 . The relative permittivity and dissipation factor increased when doped with 1.0 wt% ZnO. Oxygen vacancies are expected to be high at this doping concentration. The relative permittivity attains maximum value and dissipation factor attains minimum value when doped with 2.0 wt% ZnO. The surplus active electrons are absolutely compensated when $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics is doped with 2.0 wt% ZnO.

[Fig. 3](#page-2-0) shows the relative permittivity and dissipation factor of intrinsic and MgO-doped $Ba_{0.7}Sr_{0.3}TiO₃$ ceramics with different doping concentrations measured at 10 kHz. It is noted from [Fig. 3](#page-2-0) that the dissipation factor of $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics dramatically decreases as MgO doping concentration increases. The relative permittivity $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics increased and attained maximum value when doped with 0.5 wt% MgO. The oxygen vacancies developed when $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics is doped with MgO exists at corners of perovskite octahedra structure which are interconnected as well as movable and thus shifted to grain boundaries. This result in domain-wall pinning due to elastic as well as electric dipoles originated and hence dissipation factor is reduced in the ferroelectric state of $Ba_{0.7}Sr_{0.3}TiO₃$ ceramics [\[29\]](#page-4-0).

As we increase the doping concentration of MgO, Mg^{2+} ions at boundaries convert to MgO molecules which suppress grain growth and act as sintering agent. This is because of supremacy

Fig. 2. Relative permittivity & dissipation factor of intrinsic and ZnO-doped $Ba_{0.7}Sr_{0.3}TiO₃$ ceramics capacitors measured at 10 kHz frequency.

Fig. 3. Relative permittivity & dissipation factor of intrinsic and MgO-doped Ba_{0.7}Sr_{0.3}TiO₃ ceramics capacitors measured at 10 kHz frequency.

Fig. 4. Relative permittivity to dissipation factor ratio of intrinsic and Al_2O_3 -doped Ba_{0.7}Sr_{0.3}TiO₃ ceramics capacitors measured at 10 kHz frequency.

of domain-wall pinning with MgO molecules as pinning sites when MgO doping concentration is above solubility limit which give rise to transgranular fracture from intergranular fracture like similar ceramic composites [\[29\].](#page-4-0) This results in lowering relative permittivity of $Ba_{0.7}Sr_{0.3}TiO₃$ ceramics when doped with higher concentrations of MgO. The dissipation factor is achieved minimum value when doped with 8.0 wt% MgO. The surplus active electrons are compensated precisely in 8.0 wt% MgO-doped $Ba_{0.7}Sr_{0.3}TiO₃$ ceramics [\[16\]](#page-4-0).

As the ionic radius of the dopant ion decreases the density of doped BST ceramics decreases. This enhances porosity of BST ceramics due to inter-granular voids resulting in low relative per-mittivity [\[16\].](#page-4-0) Since the ionic radius of Al^{3+} ion is less than that of $Ti⁴⁺$, doping of Ba_{0.7}Sr_{0.3}TiO₃ ceramics with Al₂O₃ reduces relative permittivity. Thus intrinsic $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics exhibit higher relative permittivity than Al_2O_3 -doped $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics which is evident from [Fig. 1](#page-1-0).

Since the ionic radius of Zn^{2+} ion and Mg^{2+} ion are higher than that of Ti⁴⁺, doping of $Ba_{0.7}Sr_{0.3}TiO₃$ ceramics with ZnO and MgO increase the density of doped BST ceramics. This reduces

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Fig. 5. Relative permittivity to dissipation factor ratio of intrinsic and ZnO-doped $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics capacitors measured at 10 kHz frequency.

Fig. 6. Relative permittivity to dissipation factor ratio of intrinsic and MgO-doped $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics capacitors measured at 10 kHz frequency.

Fig. 7. Relative permittivity to dissipation factor ratio of intrinsic and $Al_2O_3/ZnO/$ MgO-doped $Ba_{0.7}Sr_{0.3}TiO₃$ ceramics capacitors measured at 10 kHz frequency.

Table 1

Comparison of performance parameters of proposed capacitors with related research works.

inter-granular voids and hence porosity of BST ceramics, result in higher relative permittivity with some doping concentrations of ZnO and MgO in MgO/ZnO-doped $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics than intrinsic $Ba_{0.7}Sr_{0.3}TiO₃$ ceramics which is evident from [Fig. 2](#page-1-0) and [Fig. 3](#page-2-0). Among ZnO and MgO-doped $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics, the higher value of relative permittivity is achieved for 2.0 wt% ZnOdoped $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics owing to higher ionic radius of Zn^{2+} ion than Mg²⁺ ion.

The relative permittivity increases with dissipation factor irrespective of doping material and doping concentration which is evident from [Fig. 1,](#page-1-0) [Fig. 2](#page-1-0) and [Fig. 3.](#page-2-0) Hence in order to find out the most suitable doping material and doping concentration, a figure of merit (FoM) is to be defined with the two contradicting parameters, relative permittivity and dissipation factor. Since the best ceramic material should possess highest possible relative permittivity and lowest possible dissipation factor, figure of merit can be defined as the ratio of relative permittivity to dissipation factor. The ceramics with highest figure of merit shows optimum performance.

The relative permittivity to dissipation factor ratios (FoM) of intrinsic and Al_2O_3 -doped $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics with different doping concentrations measured at 10 kHz are given in [Fig. 4](#page-2-0). Even though the relative permittivity of intrinsic $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics is higher compared to Al_2O_3 -doped $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics for all doping concentrations, the dissipation factor is also higher as shown in [Fig. 1](#page-1-0). Hence its FoM is lower compared to some of Al_2O_3 doping concentrations. It is evident from [Fig. 4](#page-2-0) that 1.0 wt% Al_2O_3 -doped $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics shows optimum performance among other doping concentrations of Al_2O_3 .

[Fig. 5](#page-2-0) shows the figure of merit of intrinsic and ZnO-doped $Ba_{0.7}Sr_{0.3}TiO₃$ ceramics with different doping concentrations measured at 10 kHz. Among these, the relative permittivity is the highest and dissipation factor is the lowest for 2.0 wt% ZnO-doped $Ba_{0.7}Sr_{0.3}TiO₃$ ceramics [\(Fig. 2](#page-1-0)). Hence its FoM is the highest and shows optimum performance among other doping concentrations of ZnO as given in [Fig. 5.](#page-2-0)

[Fig. 6](#page-2-0) shows the FoM of intrinsic and MgO-doped $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics for different doping concentrations measured at 10 kHz. Even though the relative permittivity is the highest for 0.5 wt% MgO-doped $Ba_{0.7}Sr_{0.3}TiO₃$ ceramics, its dissipation factor is not low enough to show optimum performance as shown in [Fig. 3.](#page-2-0) From [Fig. 6,](#page-2-0) it is seen that FoM of 4.0 wt% MgO-doped $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics is the highest and shows optimum performance among other doping concentrations of MgO.

The dissipation factor achieved by 1.0 wt% Al_2O_3 , 2.0 wt% ZnO and 4.0 wt% MgO doped $Ba_{0.7}Sr_{0.3}TiO₃$ ceramics are 0.0189, 0.0087 and 0.0116 respectively. The dissipation factor achieved by 2.0 wt% ZnO is promisingly very low. [Fig. 7](#page-2-0) shows the relative permittivity to dissipation factor ratio (figure of merit) of intrinsic

and $Al_2O_3/ZnO/MgO$ -doped $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics with different doping concentrations measured at 10 kHz for comparing the performance of capacitors with all doping materials and doping concentrations. It is evident from [Fig. 7](#page-2-0) that among the doping materials and doping concentrations considered in this work, 2.0 wt% ZnO-doped $Ba_{0.7}Sr_{0.3}TiO₃$ ceramics possesses highest figure of merit and hence optimum performance.

The performance parameters such as relative permittivity, dissipation factor of proposed $Al_2O_3/ZnO/MgO$ -doped $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics based MIM capacitors are compared with related research works and is given in Table 1. The doping concentrations of dopant materials are also compared. Some researchers added dopant materials in mol% and some others in wt%. In this work, the dopant materials were added in wt% of insulator material $Ba_{0.7}Sr_{0.3}TiO_3.$

The 2.0 wt% ZnO-doped $Ba_{0.7}Sr_{0.3}TiO₃$ ceramics based MIM capacitor proposed in this work shows better relative permittivity compared to the recent research works. The 4.0 wt% MgO-doped $Ba_{0.7}Sr_{0.3}TiO₃$ ceramics based MIM capacitor proposed in this work also shows better relative permittivity compared to the recent research works except capacitors proposed by Liang et al. [\[16\]](#page-4-0) and Laishram et al. [\[35\]](#page-4-0).

4. Conclusion

The dielectric properties of $Ba_{0.7}Sr_{0.3}TiO₃$ ceramics when doped with Al_2O_3 , ZnO and MgO were investigated. The relative permittivity and dissipation factor of intrinsic $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics and $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics doped with 0.5, 1.0, 2.0, 4.0 and 8.0 wt% of all the doping materials, Al_2O_3 , ZnO and MgO were measured at room temperature. The relative permittivity increases with dissipation factor irrespective of doping material and doping concentration. The dissipation factor achieved by 2.0 wt% ZnO doped $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics is 0.0087, which is promisingly very low. Further it was observed that 2.0 wt% ZnO doped $Ba_{0.7}Sr_{0.3}TiO₃$ ceramics possesses highest figure of merit (relative permittivity to dissipation factor ratio) and hence optimum performance among the doping materials and doping concentrations considered in this paper. The performance parameters of proposed $Al_2O_3/ZnO/MgO$ doped $Ba_{0.7}Sr_{0.3}TiO₃$ ceramics based MIM capacitors were compared with related research works as well.

CRediT authorship contribution statement

P.S. Smitha: Conceptualization, Data curation, Investigation, Writing - original draft. **Jitha S. Jayan:** Methodology, Resources. Appukuttan Saritha: Formal analysis, Validation. V. Suresh Babu:

Project administration, Writing - review & editing. G. Shiny: Supervision, Writing - review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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