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Investigation on dielectric properties of Al₂O₃, 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 Ba0,7Sr0,3TiO3 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% Al₂O_{3.} 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}TiO₃ 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}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. © 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], phase shifter application [7], energy storage application [8] and power application [9]. Hence it is very crucial to reduce the dissipation factor of BaSrTiO₃ ceramics materials [10]. 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], Ti [13], La [14], W [15], Al [16], Ce [17], Cr [18], Bi [19], Mg [20], Zr [21], Mn [22] and Fe [23] to lower the dissipation factor.

In this work, dielectric properties of Ba_{0.7}Sr_{0.3}TiO₃ ceramics when doped with Al₂O₃, ZnO and MgO are investigated. The dopant solubility in Ba0.7Sr0.3TiO3 ceramics depends mainly on its ionic radius [24]. 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]. The ionic radii of Al³⁺, Zn²⁺ and Mg²⁺ ions are 0.675 A°, 0.88 A° and 0.86 A° respectively [27] which are comparable to that of Ti⁴⁺. This enhances the chances for Al³⁺, Zn²⁺ and Mg²⁺ 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 °C respectively which are well below the sintering temperature of BST samples (1275 °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 SrCO₃ 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° 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 Ba0.7Sr0.3TiO3 nanopowder are 0.7478 g and 0.2522 g respectively.

The synthesized Ba_{0.7}Sr_{0.3}TiO₃ sample was blended with 0.5, 1.0, 2.0, 4.0 and 8.0 wt% Al₂O₃ (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 °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 $Ba_{0.7}Sr_{0.3}TiO_3$ 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 $Ba_{0.7}Sr_{0.3}TiO_3$ 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]. 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] at same frequency and temperature.

The relative permittivity and dissipation factor of intrinsic and ZnO-doped Ba_{0.7}Sr_{0.3}TiO₃ 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 Al₂O₃. 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}TiO₃ ceramics is doped with 2.0 wt% ZnO.

Fig. 3 shows the relative permittivity and dissipation factor of intrinsic and MgO-doped $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics with different doping concentrations measured at 10 kHz. It is noted from Fig. 3 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 $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics [29].

As we increase the doping concentration of MgO, Mg²⁺ ions at boundaries convert to MgO molecules which suppress grain growth and act as sintering agent. This is because of supremacy



Fig. 1. Relative permittivity & dissipation factor of intrinsic and Al₂O₃-doped Ba_{0.7}Sr_{0.3}TiO₃ ceramics capacitors measured at 10 kHz frequency.



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.

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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 $Ba_{0.7}Sr_{0.3}TiO_3$ 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]. 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].

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 permittivity [16]. Since the ionic radius of Al^{3+} ion is less than that of Ti⁴⁺, doping of $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics with Al_2O_3 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.

Since the ionic radius of Zn^{2+} ion and Mg^{2+} ion are higher than that of Ti^{4+} , doping of $Ba_{0.7}Sr_{0.3}TiO_3$ 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}TiO₃ 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 $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics capacitors measured at 10 kHz frequency.

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Table 1

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

Reference		Insulator material	Dopant material	Doping concentration	Relative permittivity	Dissipation factor
Lee et al. (2002)	[20]	Ba _{0.7} Sr _{0.3} TiO ₃	MgO	5 mol%	372	0.0037
Cole et al. (2002)	[30]	Ba _{0.6} Sr _{0.4} TiO ₃	Mg acetate	1 mol%	423	0.009
Liang et al. (2003)	[16]	Ba _{0.6} Sr _{0.4} TiO ₃	Al_2O_3	0.8 wt%	2500	0.0015
Chong et al. (2004)	[4]	Ba _{0.5} Sr _{0.5} TiO ₃	Al_2O_3	9 0.0 wt%	870	0.011
Hu et al. (2005)	[31]	Ba _{0.5} Sr _{0.5} TiO ₃	MgO	20 wt%	450	0.0002
Dong et al. (2009)	[32]	Ba _{0.3} Sr _{0.7} TiO ₃	ZnO	1.6 wt%	610	0.00025
Ham et al. (2011)	[33]	Ba _{0.5} Sr _{0.5} TiO ₃	LiCO ₃	1 wt%	1441	0.01
Chou et al. (2012)	[34]	Na _{0.5} Bi _{0.5} TiO ₃	ZnO	0.5 wt%	216	0.133
Laisharam et al. (2016)	[35]	Ba _{0.7} Sr _{0.3} TiO ₃	MgO	10 mol%	2037	0.007
Lu et al. (2017)	[36]	0.8BaTiO ₃ - 0.2Bi(Zn _{1/2} Ti _{1/2})O ₃	Nb ₂ O ₅	0.5 wt%	1580	0.021
Hossain et al. (2018)	[37]	BaTiO ₃	NiO	0.5 wt%	1050	0.05
Panigrahi et al. (2019)	[38]	BiFeO ₃ -BaTiO ₃	Ga_2O_3	1.4 mol%	500	Not given
This work		Ba _{0.7} Sr _{0.3} TiO ₃	Al_2O_3	1.0 wt%	1227	0.0189
			ZnO	2.0 wt%	3152	0.0087
			MgO	4.0 wt%	2035	0.0116

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 $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics which is evident from Fig. 2 and Fig. 3. 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% ZnO-doped $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics owing to higher ionic radius of ZnO²⁺ ion than Mg²⁺ ion.

The relative permittivity increases with dissipation factor irrespective of doping material and doping concentration which is evident from Fig. 1, Fig. 2 and Fig. 3. 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. 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. Hence its FoM is lower compared to some of Al_2O_3 -doped $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics. It is evident from Fig. 4 that 1.0 wt% Al_2O_3 -doped $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics among other doping concentrations of Al_2O_3 .

Fig. 5 shows the figure of merit of intrinsic and ZnO-doped $Ba_{0.7}Sr_{0.3}TiO_3$ 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 $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics (Fig. 2). Hence its FoM is the highest and shows optimum performance among other doping concentrations of ZnO as given in Fig. 5.

Fig. 6 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 $Ba_{0.7}Sr_{0.3}TiO_3$ ceramics, its dissipation factor is not low enough to show optimum performance as shown in Fig. 3. From Fig. 6, 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 $Ba_{0.7}Sr_{0.3}TiO_3$ 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 shows the relative permittivity to dissipation factor ratio (figure of merit) of intrinsic

and Al₂O₃/ZnO/MgO-doped Ba_{0.7}Sr_{0.3}TiO₃ 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 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] and Laishram et al. [35].

4. Conclusion

The dielectric properties of Ba_{0.7}Sr_{0.3}TiO₃ ceramics when doped with Al₂O₃, ZnO and MgO were investigated. The relative permittivity and dissipation factor of intrinsic Ba0.7Sr0.3TiO3 ceramics and Ba0.7Sr0.3TiO3 ceramics doped with 0.5, 1.0, 2.0, 4.0 and 8.0 wt% of all the doping materials, Al₂O₃, 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}TiO₃ 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 Al₂O₃/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:** P.S. Smitha, J. S. Jayan, A. Saritha et al.

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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