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Structural and Dielectric Properties of $\text{Sr}_2\text{TiMnO}_6$ Ceramic and Polymer Composites: Review

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ABSTRACT: Application of polymer/ceramic composites in the electronics and automotive industries are on the rise, due to their improved efficiency and design properties. This review study comprehensively focuses on the primary processing technique, structural, dielectric permittivity and dielectric loss. It is clear that there is always an increase in dielectric constant (ϵ_r) for an increase in volume fraction or weight percentage in case of ceramic-polymer composites.

KEYWORDS: Ceramic, Polymer, Double perovskites, Dielectric constant.

I. INTRODUCTION

Due to their peculiar electrical and magnetic properties, double perovskites have recently attracted a lot of interest. The stoichiometric formula for double perovskites is $\text{A}_2\text{B}^{\prime}\text{B}^{\prime\prime}\text{O}_6$. Alkaline earth metals such as Ca, Ba, Sr, and others, or rare earth ions with greater ionic radii, occupy the A site. The transition metal cations with smaller ionic radii occupy the B^{\prime} and $\text{B}^{\prime\prime}$ octahedral sites. Depending on the degree of octahedral tilting that changes the extent of orbital overlap between the B-site octahedral networks [1, 2], double perovskite exhibits an interesting combination of conductivity, dielectric, and magnetic properties. For their relatively low K dielectric properties, double perovskite oxides such as LaBaZnTaO_6 , LaBaZnNbO_6 , and $\text{Ba}_2\text{Zn}_{0.5}\text{Ti}_{0.5}\text{TaO}_6$ have been studied [3]. The magneto dielectric effect in ferromagnetic semiconductors such as $\text{La}_2\text{NiMnO}_6$ and $\text{La}_2\text{MnCoO}_6$ near room temperature has provided a new perspective on the search for the coexistence of dielectric and magnetic properties in related compounds [4-6]. Due to their remarkable aspects such as dielectric and magnetic properties, generic formula compounds Sr_2MMnO_6 (where M is a diamagnetic transition metal cation such as Sb^{5+} , Ti^{5+} , Nb^{5+} , Mo^{6+} , Ta^{5+} , etc) is being studied extensively [7-10].

Roa-Rojas et al. were the first to describe the structure and magnetic properties of $\text{Sr}_2\text{TiMnO}_6$ (STMO) ceramics [11]. According to this paper, STMO has a tetragonal structure with lattice parameters of $a=5.4702 \text{ \AA}$ and $c= 7.7361 \text{ \AA}$, as well as a ferromagnetic to paramagnetic transition at 44.8 K. STMO ceramics were previously reported to have high dielectric constant values, which were attributed to Maxwell-Wagner type relaxation behavior (tests were only conducted in a small temperature range of 180 to 300K). Materials which possess high dielectric constant i.e., $>10^3$ have gained more technological importance because of their various applications in the areas of microelectronics such as capacitors, memory devices [12]. Also, high permittivity materials can be used to reduce the size of electronic equipment. These capacitors make up a large part of electrical and electronic circuits, and their primary function is to store electrical energy.

Polymers are used as dielectrics in capacitors because of their high breakdown strength and low dielectric loss. However, the polymers fundamental drawback is their low dielectric permittivity (2-3). Ceramics, on the other hand, have a higher permittivity than polymers, but their fundamental disadvantage is their low breakdown strength and significant dielectric loss [13]. The creation of hybrid materials like polymer/ceramic composites combines the advantages of polymer and ceramics. Composites can aid in the miniaturization of electronic devices because to their higher dielectric permittivity than polymers [14]. Both the softness of the polymer and the advantages in electric characteristics would be created by combining ceramic and polymer.

This study examines high dielectric constant composites for dielectric application, as well as research accomplishments and advancements in the production of polymer/ceramic composites.

II. METHODOLOGY

In this study, the research papers selected are having $\text{Sr}_2\text{TiMnO}_6$ ceramics, which possess double perovskites in nature. The various authors discussed the solid-state reaction method of preparation in their respective papers. The method

followed for synthesizing STMO ceramics is conventional solid-state reaction route. The stoichiometric amounts of precursors SrCO_3 , TiO_2 and MnO_2 were mixed according to the chemical formula $\text{Sr}_2\text{TiMnO}_6$. Mixture was thoroughly mixed using alcohol/acetone medium and ball milled (300rpm) for 3 hr. The resulting homogeneous mixture is dried for 1 hour before being transferred to a recrystallized alumina crucible and calcined in three stages at 1000°C , 1200°C , and 1370°C to remove all carbonates to get phase pure STMO powder [14-17]. The existence of distinctive peaks of the double perovskite system is revealed by XRD analysis [11].

In this review paper, the research papers are also selected which are having different polymer and STMO ceramic composites [15-16]. The method followed for synthesizing the composites is melt-mixing technique. $(cd/\epsilon A)$ was used to compute the dielectric constant (ϵ_r), where 'd' is the thickness of the film or pellet and 'A' is the area of the produced pellet.

III. RESULTS

Structural properties

Double perovskites have stoichiometric formula $\text{A}_2\text{B}'\text{B}''\text{O}_6$, where the B-sites, i.e., B', B'' can be occupied by the transition metals [18, 19]. The crystal structure and space group of double perovskites are determined by the arrangement of $\text{B}'\text{O}_6$ and $\text{B}''\text{O}_6$ octahedra, specifically ordering and tilting [20]. In 3-dimension crystal structure, there are three types of B-site cationic arrangements: round, rock salt, and layered arrangements [19]. As seen schematically, randomly ordered octahedral is typically $\text{pm}3\text{m}$ [Fig 1]. A site (0,0,0) divalent cations occupy the corner locations in the unit cell of randomly arranged double perovskite. Tetravalent cations from B-sites (B' and B'') are seated in the body-centered position ($\frac{1}{2}, \frac{1}{2}, \frac{1}{2}$), while oxygen anions are seated in the face-centered position ($\frac{1}{2}, \frac{1}{2}, 0$). $\text{B}'\text{O}_6$ and $\text{B}''\text{O}_6$ octahedra are organized alternatively in three-dimensional space in a double perovskite structure with rock salt ordering, resulting in distinct space groups such as $\text{Fm}3\text{M}$, $\text{I}4/\text{m}$, and $\text{P}4/\text{nmc}$. The layered kind of double perovskite is the least common structure, in which the layers of $\text{B}'\text{O}_6$ and $\text{B}''\text{O}_6$ octahedra are alternately oriented only in one direction.

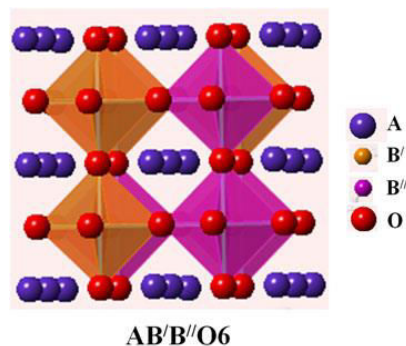
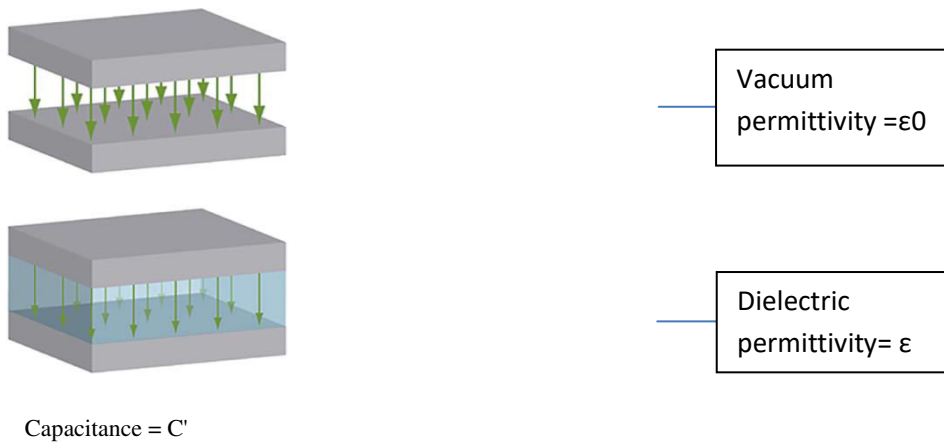


Fig.1. Schematic of $\text{A}_2\text{B}'\text{B}''\text{O}_6$ double perovskite structure

Dielectric permittivity

Due to polarization, the dielectric constant decreases with increasing frequency at all temperature [21]. The presence of electrode polarization effect can explain the high dielectric constant values. The capacitance of a capacitor filled with the supplied material to the capacitance of an identical capacitor in a vacuum without dielectric material equals ϵ_r . When a dielectric is inserted between the plates of a parallel-plate capacitor, the capacitance, or ability to store opposite charges on each plate, is always increased, compared to when the plates are separated by a vacuum [22]. C' is the capacitance of a capacitor filled with a given dielectric, and C is the capacitance of a capacitor filled with the same dielectric in a vacuum (Fig.2). The ϵ_r is a dimensionless number that can be written as $\epsilon_r = \frac{C'}{C}$



Capacitance = C'

$$\epsilon_r = C'/C \quad \epsilon_r = \epsilon/\epsilon_0$$

Fig.2. Dielectric permittivity concept

Increase in dielectric constant value with an increase in volume fraction which $\tan\delta$ decreases for dielectric characteristics of composites. The outcome is attributable to an increase of ceramic particle reinforcement in the polymer matrix, which has a higher dielectric constant. The dielectric constant in polymer composites has been reported to rise with ceramic volume percentage, and there is a dielectric constant value difference between the polymers, which is due to polymer relaxing with ceramic [23].

To rationalize the electrode influence, interfacial polarization on the dielectric behavior of STMO, dielectric constant measurements were done with several electrode materials. At room temperature, three distinct electrodes are used to present dielectric constant as a function of frequency: silver paints, sputtered silver, and sputtered gold. At low frequencies, high dielectric constant (ϵ_s) values are attained in all three instances. However, the resistance of sputter electrodes (gold and silver) is significantly larger than that of silver paint electrodes. This is owing to the good wettability of the sputtered nano sized metal clusters on the surface. The silver paint, on the other hand, has very large particles [17]. Other parameters include the work function of the electrode material and the thickness of the Schottky barrier. On the dielectric behavior of STMO, the work functions of sputtered gold and silver electrodes are nearly identical.

Sample thickness dependent dielectric measurements are carried out at room temperature using sputtered gold as the electroding material in the [K. R. S. PreethiMeher et al.] study report. The dielectric constant (ϵ_s) of thicker samples is much higher (at low frequencies). While ϵ_∞ is independent of the thickness of the sample [17]. In the low frequency region, the dielectric constant is $\sim 10^4$ at room temperature. When the temperature was raised, the value of the dielectric constant decreased [22]. With increasing frequencies, the dielectric changes to higher temperatures, accompanied by a peak in the $\tan\delta$ curve. The size of the step for 300Hz is ~ 5000 , which is less than the $\sim 10^5$ stated for 1 KHz in [17]. This reveals that the preparation method has influence on the dielectric characteristics of STMO. It also indicates that Mn cation decreases the polarizability of STMO lattice and suppresses the ferroelectric characteristics [24].

In the low frequency band of 1 Hz- 1KHz, the permittivity of ϵ_r of pure PTFE and PTFE/STMO composites was discovered to be evident [25]. As the STMO content in the PTFE matrix increased, ϵ_r increased as well. ϵ_r was obtained to be at 2.58 for pure PTFE and increased to 4.24 when 15 wt% STMO was added [15]. The inter particle distance in the PMMA matrix decreases as the STMO content increases. Because of the overlapped diffused electrical double layer around each particle, PMMA/STMO composites have a higher dielectric constant [16]. The composites was able to obtain a permittivity of 30.9 @ 100Hz.

Dielectric loss

An alternating electric field causes tangent δ ($\tan\delta$) or energy loss in a dielectric. It is a material attributed of a capacitor rather than a geometrical property. In general, the tangent ($\tan\delta$) expressed as a loss tangent ($\tan\delta$) can be mathematically defined as



$$\tan\delta = \frac{\epsilon''}{\epsilon'}$$

Where, angle δ is supplementary angle of the phase difference between the applied electric field and induced current.

ϵ'' and ϵ' are the real and imaginary parts of the ϵ_r .

As the frequency lowers, the dielectric loss increases to larger values, indicating the Maxwell-Wagner (MW) effect [21]. Dielectric loss in pure and composite materials reveals the materials capacitor performance, with significant dielectric loss indicating dissipation of stored energy in the form of heat [16]. With the addition of STMO, the dielectric loss of PTFE was also increased, because of the conduction losses caused by the presence of STMO in the composites [15]. At 100 Hz, the loss values increased from 0.00016 to 0.21835 for pure and 15 wt% composites.

IV. CONCLUSION

The solid state reaction approach was used to make a variety of double perovskite $\text{Sr}_2\text{TiMnO}_6$ based polymer composites in this study. These double perovskites have a dense microstructure with grain size distribution scaling from nm to μm . These investigations have demonstrated the promise of STMO for dielectric applications with high dielectric permittivity (ϵ_r) and low $\tan\delta$ values. The thickness of the sample, the temperature, and the matrix all influence dielectric property tests. These materials are ideal for use in capacitors.

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