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Giant effect on structural, magnetic, electrical, and optical properties of lead-free Ba$_{0.6}$Sr$_{0.4}$Ti$_{1-x}$Al$_x$O$_3$ ceramics via Sr and Al Co-doping engineering

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Abstract

In this research, structural, magnetic, electrical, and optical properties of $A$-substituted Ba$_{0.6}$Sr$_{0.4}$Ti$_{1-x}$Al$_x$O$_3$ ceramics (where $x = 0.00$ to $0.20$) have been investigated and discussed. The ceramic samples have been successfully synthesized by the conventional solid-state reaction method. The crystallinity of the prepared samples has been confirmed by x-ray peak broadening techniques such as the Williamson Hall (W-H) plot technique and the Scherrer method. The lattice constant $a'$, showing a significant drop as Al concentration increases, varies from 3.942 Å to 3.921 Å. The FTIR spectrum revealed a prominent peak between 435 cm$^{-1}$ to 540 cm$^{-1}$, without showing any secondary phase. Raman spectroscopic analysis showed that the prepared samples are in cubic phase with no phase transition. The shifting of a dominating peak in Raman at 580 cm$^{-1}$ is due to the development of Al$_{2}$Ti defects. VSM analysis at room temperature showed the ferromagnetic characteristics of all the samples. The highest value of the anisotropic constant (Ka $\sim$ 39.933 emu cm$^{-3}$) has been seen at $x = 0.10$. The real component of initial permeability is almost constant for the frequency range of about 75 MHz, and above this frequency range, it quickly reduces. At the high-frequency range, the prepared samples show an excellent relative quality factor due to the low dielectric loss, notably when $x = 0.05$. The prepared perovskites exhibited minimum eddy current loss characteristics due to the increment of AC resistivity at $x = 0.20$. UV–visible spectroscopy shows a significant change in the energy bandgap from 3.23 eV to 3.04 eV with the doping concentrations.

1. Introduction

Recently, ferroelectric ceramics have garnered much interest because of their simplicity of manufacture and ability to acquire different desirable characteristics via compositional changes [1]. Ferroelectric crystals demonstrate stable and switchable electric polarization by accommodating atomic displacements [2]. When comparing other lead-based ferroelectric materials to BaTiO$_3$ (BTO)-based ceramics, it has been determined that BaTiO$_3$ (BTO) based ceramics are more appealing choices for this feature because of their simple crystal structure, low leakage current, exceptionally high dielectric constant, anisotropic optical behavior, and excellent stability [3–5]. It is a potential choice for microelectronic and electro-ceramic systems such as capacitors, piezoelectric devices, semiconductors, thermistors, ultrasonic transducers, and high storage memory devices for these greater piezoelectric, ferroelectric, and dielectric performance [6].

Furthermore, because of its capacity to retain several differentials, concurrently its research is being pursued on a large scale. Additionally, depending on the processing method and synthesis route, dopant inclusion may significantly alter its characteristics [7]. Doping the original state of BaTiO$_3$ converts it from an insulator to a semiconductor [8]. BaTiO$_3$ may be doped to form a double perovskite with dopant on both sites (A-site and B-site), resulting in dramatic changes in its dielectric and magnetic characteristics at standard room temperature, as well as the ability to exhibit multiferroicity [9]. For example, Ba$_{1.5}$Sr$_x$TiO$_3$ (BST) perovskite...
developed from BaTiO$_3$ is one of the most significant ABO$_3$-type ferroelectrics (BT) materials. Because of its composition and ferroelectric characteristics associated with Curie temperature, barium strontium titanate (BST, Ba$_{1-x}$Sr$_x$TiO$_3$), a versatile material, has piqued the attention of engineers and researchers recently [10, 11]. BST has been successfully used in various applications, including pressure transducers, sensors, high-voltage capacitors, detectors, piezoelectric, tunable filters, optoelectronic devices, and actuators [12]. The electrical characteristics of BST were altered when Sr$^{2+}$ replaced Ba$^{2+}$ in origin BT. The compositional design, microstructure, and grain size all affect the electrical characteristics of BaTiO$_3$ besides the doping technique [13]. It is now being investigated extensively for a range of sensor applications, including pyroelectric, piezoelectric, gas, and humidity sensing [14].

Numerous methods have been used to synthesize BST nanostructures, including hydrothermal approaches [15], sol-gel [16], Chemical deposition [17], ball milling [18], Solvothermal [19], co-precipitation [20], Neutron diffraction [21], Tartrate precursor [22], Blinder jetting [23], and solid-state technique [24]. Researchers have experimented with BT and BST in the presence of a variety of dopants, including Bi [25], Ca [26], Dy [27], Er [28], Eu [29], Fe [30], Ho [31], Hf [32], La [33], Nb [34], Rh [35], Sb [36], Sn [37], V [18], Y [38], Zn [39], and Zr [40]. Some existing ceramics have been developed using strontium doped with barium titanate [41–43]. Some have been developed using aluminum doped with barium titanate [44]. However, these existing ceramics are unable to give all desired properties, especially structural properties by Raman and FTIR, magnetic properties, and optical properties. There are several challenges to be developed.

Among all possible BST compositions, Ba$_{0.6}$Sr$_{0.4}$TiO$_3$ was selected as the parent material due to its favorable combination of low thermal conductivity, high dielectric constant, relatively low loss tangent, and acceptable tenability [45, 46].

According to the study, doping Al with BaTiO$_3$ enhanced the dielectric constant, lowered the Curie temperature, and relocated the dielectric loss peaks to a higher frequency [47]. An Al$^{3+}$ substitution may occupy the $T$ site in the BaTiO$_3$ lattice, although it needs a charge as compensation [48]. As a result of this charge compensation process, oxygen vacancies are generated, which might explain the presence of Al$^{3+}$ (ionic radius of Al$^{3+}$, $r = 1.84$ Å) at the BTO perovskite’s $T$ site (B-site, ionic radius of Ti$^{4+}$, $r = 0.6$ Å) [49]. Thus, the presence of oxygen vacancies in the composition causes semiconducting behavior, as shown by the equation [50].

\[
O^{-2} = \frac{1}{2} O_2 + 2e^{-}
\]  

Consequently, Al$^{3+}$ doping on the B site of the BTO perovskite increases the magnetic properties of the material. Due to differences in the magnetic moments of Al$^{3+}$ and Ti$^{4+}$ as well as the ionic radii between these cations, a significant lattice distortion will arise at the B site, which will increase magnetic characteristics [51, 52].

At the B site, which provides the improved multiferroic perovskite characteristics. The novelty of this research work is that the structural, magnetic, electrical, and optical properties of Al$^{3+}$ substituted BSTAO perovskites composition of Ba$_{0.6}$Sr$_{0.4}$Ti$_{1-x}$Al$_x$O$_3$ have not yet been done according to the literature review. In this research, we have investigated the barium titanate co-doped with strontium and aluminum, where 40% strontium was used at the A-site in Ba$_{0.6}$Sr$_{0.4}$TiO$_3$ ceramic as the parent composition. Here, Ba (an oxidation state of +2) was substituted with Sr (an oxidation state of +2) at the A-site, with Ba$_{0.6}$Sr$_{0.4}$TiO$_3$ serving as the elemental composition where Al was doped in 0.05 molar increments. Thus, we sought to ascertain the effect of Al doping (at the B-site) when Sr is maintained at a constant ratio (molar ratio of 0.4) at the A-site.

In this study, Al$^{3+}$ substituted composition of Ba$_{0.6}$Sr$_{0.4}$Ti$_{1-x}$Al$_x$O$_3$ perovskites have been investigated with the hope of having better structural, morphological, magnetic, and electrical properties to fulfill the following objectives.

- To investigate the characteristics of Sr and Al co-doped BSTAO perovskites composition of Ba$_{0.6}$Sr$_{0.4}$Ti$_{1-x}$Al$_x$O$_3$ for numerous industrial applications, including non-volatile ferroelectric FRAM’s memories, multilayer Ceramic (MLCCs), gaseous pollutant detection like CO, optical data storage at high-density, positive temperature coefficient resistors (PTCR), energy storage devices, ultrasonic transducer, thermistors, and capacitors for future applications in electro-ceramics and advanced electronics.
- To obtain low dielectric loss and high dielectric constant which make the ceramic compositions one of the attractive choices for tunable microwave device applications and dynamic random-access memory (DRAM).
- To reduce the optical band gap energy of the ceramic compositions for optoelectronic applications.

Finally, the limitations of this research are discussed in detail here. Due to intra-granular pores and grain impurity, specific suppressed points on the compositions’ surface cause a drop of magnetization at the high-
frequency region. Moreover, the magnetization of this Ba$_{0.6}$Sr$_{0.4}$Ti$_{1-x}$Al$_x$O$_3$ perovskite composition is affected by several parameters, including composition, stoichiometry, grain size, coercive field, crystal anisotropy, porosity, and impurity contents. The existence of a diamagnetic component such as BaTiO$_3$ is typically related to a decrease in magnetization at high fields. Besides, the magnetic anisotropy and a tiny number of impurities, i.e., aluminum oxide or strontium oxide, in the composition may also explain the anomaly in magnetization. The presence of porosity in the prepared samples, which may be developed during sintering, can induce variations in bonding among the composition’s components, resulting in a shift in the interatomic distance, which affects the density of the compositions. Compositional inhomogeneities produce interfacial defects, grain structure, impurities, and porosity, contributing to interfacial polarization, which affects the dielectric constant. These inhomogeneities are introduced into the ceramics during pre-sintering and high-temperature sintering operations. Furthermore, the dielectric constant is influenced by various parameters, including sintering temperature, stoichiometry, structural homogeneity, and compositional performance of the ceramics.

2. Experimental details

2.1. Sample preparation

The conventional solid-state reaction technique was used to synthesize polycrystalline Ba$_{0.6}$Sr$_{0.4}$Ti$_{1-x}$Al$_x$O$_3$ samples where x = 0.00, 0.05, 0.10, 0.15, and 0.20 respectively. The starting raw materials of very pure grade, strontium oxide (SrO, 99.99%), barium carbonate (BaCO$_3$, 99.99%), alumina (Al$_2$O$_3$, 99.99%), and titanium dioxide (TiO$_2$, 99.99%) were used to synthesize the proper perovskite compositions. Firstly, all of these basic components were weighed and combined to a stoichiometric ratio. To obtain the desired compound, these components were ground with a pestle and mortar for approximately six hours. Then these powders of compositions were treated into a disc shape for pre-sintering at 700°C for four hours to eliminate volatile contamination. These samples were again milled for two hours after pre-sintering. Next, the powders of all samples were mixed with a binding agent polyvinyl alcohol (PVA) of 10 wt%. After that, the powders were pressed under 45 MPa pressure using a hydraulic press. Both cylinder-shape and disk-shaped samples (five samples of each type of ceramic were made) were produced for the characterization at 10 and 15 kN m$^{-2}$ units. Finally, the samples of different compositions were sintered in the presence of air at 1250°C for 4 h [53–56].

2.2. Characterization technique

To investigate the structural analysis and phase formation of the synthesized composites, an x-ray diffractometer (Philips X’Pert Pro) was used to determine XRD patterns with Cu-Kα radiation ($\lambda = 1.5406$ Å) at a 10/min rate of scanning speed at room temperature with 40 KV at 30 mA in the $2\theta$ range of 20°–70° and 0.02° steps. From the XRD data, the following relation was used to compute the lattice parameters of the prepared samples.

$$\frac{1}{d_{hkl}^2} = h^2 + k^2 + l^2 \quad \text{(for cubic)}$$

In this equation, h, k, and l denote the crystal plane’s Miller indices. The Scherrer equation was used to compute crystallite sizes in nanometers.

$$D = \frac{k \lambda}{\beta \cos \theta}$$

Where $\beta$ denotes FWHM (full width at half maximum) in radians, k denotes the broadening constant, and $\theta$ represents the diffracting Bragg’s angle in degrees.

The Williamson-Hall method can be employed to quantify the widening of the XRD diffraction patterns caused by lattice strain and crystallite size, which are both independent. The subsequent relations were applied to detect the widening:

$$\beta = \beta_{\text{crystalline}} + \beta_{\text{strain}}$$

$$\beta \cos \theta = \frac{k \lambda}{D} + 4\varepsilon \sin \theta$$

Where $\beta$ denotes the peak broadening (FWHM), $\varepsilon$ denotes the lattice strain, and D indicates the crystallite size. Here,

$$\beta_{\text{crystalline}} = \frac{x \lambda}{D \cos \theta}$$

$$\beta_{\text{Strain}} = 4\varepsilon \tan \theta$$
The following equation was used to figure out how much micro-strain was generated by doping:
\[ \varepsilon = \frac{\beta \cdot \cos \theta}{4} \] (8)

The undermentioned formula was used to compute x-ray density (theoretical density):
\[ \rho_{th} = \frac{ZM}{N.N} \] (9)

Where M, V, N_A, and Z represent the molecular mass, the unit cell volume, the Avogadros’ number, and the number of molecules per unit cell (for perovskite material, Z = 1), separately. The experimental density of all samples was calculated using the subsequent equation.
\[ \rho_{exp} = \frac{m}{V} = \frac{m}{\pi r^2 h} \] (10)

Where V denotes the volume, m represents the mass, h represents the thickness, and r denotes the radius of the expected cylindrical pellets.

The following formula was used to determine the porosity (P) of the prepared samples:
\[ P(\%) = \left[ \left( \rho_x - \rho_h \right) / \rho_x \right] \times 100\% \] (11)

The undermentioned relation was used to determine the dislocation of all the samples:
\[ \delta = \frac{1}{D^2} \] (12)

The Raman spectra of the prepared materials were captured using a Confocal MonoVista CRS+ Raman Microscope equipped with a 533 nm laser excitation source at standard room temperature and a resolution of about 1 cm\(^{-1}\). The IR transmittance spectra of samples heat-treated at 1250 °C for 4 h were collected using an FTIR Perkin-Elmer (2000) in the 4000–200 cm\(^{-1}\) range. The KBr pelleting technique was applied to analyze the samples. UV–vis spectroscopy was performed using a UV–vis spectrophotometer from Perkin Elmer (UV-2450).

The dielectric evaluation was carried out using an impedance Analyzer from WAYNE-KERR 6500B at standard room temperature by applying the silver paste on both surfaces of the Ceramic pellets to improve electrical conductivity.

The relative dielectric constant is a complex quantity comprising two variables: real and imaginary. i.e.
\[ \varepsilon^* = \varepsilon' + j\varepsilon'' \] (13)

Where, \( \varepsilon'' \) and \( \varepsilon' \) represent the imaginary component and real component of the dielectric constant, respectively. The next formula was used to determine the real component:
\[ \varepsilon' = \frac{Cd}{\varepsilon_0 A} \] (14)

Where, \( \varepsilon_0 = \) permittivity in free space = \( 8.854 \times 10^{-12} \) F m\(^{-1}\), A denotes the electrode’s cross-sectional area, and c denotes the capacitance of the sample.

The imaginary component was calculated by the subsequent formula:
\[ \varepsilon'' = \varepsilon' \tan(\delta) \] (15)

Where tan (\( \delta \)) represents loss factor in a dielectric material. The imaginary component and real component of the dielectric constant are used to calculate the loss factor:
\[ \tan \delta = \frac{\varepsilon''}{\varepsilon'} \] (16)

The following formula is used to compute the resistivity of the samples:
\[ \rho = \frac{RA}{l} \] (17)

Where \( l, A, \) and \( R \) denote the thickness, the surface area of the pellets, and the resistance of the ceramic, respectively.

Each sample’s magnetizations versus applied magnetic field curves (M–H hysteresis) were measured using a Quantum Design PPMS DynaCool instrument. Using an Impedance Analyzer from Wayne-Kerr (series 6500B), the complex permeability of the prepared compositions was calculated within the range of 10\(^5\) Hz–10\(^8\) Hz. The complex permeability’s real component \( \mu_r \) and imaginary component \( \mu_i'' \) were measured using the following equations:
where $\mu''_t = \mu'_t \cdot \tan \delta$.

And, $L_0$ denotes the winding coil’s inductance when the sample core is absent, and $L_S$ represents the self-inductance of the ceramic core. And, $L_0$ is deduced from this formula:

$$L_0 = \frac{\mu''_t N^2 S}{\pi d}$$

In the expression, $S = ((d_1 - d_2) \times h)$ and $d = \frac{d_1 + d_2}{2}$, Where $d_2$, $d_1$, and $h$ represent the outer diameter, inner diameter, and thickness of the sample separately.

### 3. Results and discussions

#### 3.1. Structural characterization

XRD is essential for observing the atomic crystal structure and determining unit cell characteristics. Our research effort has characterized all compositions sintered at 1250 °C temperature. The XRD patterns of all ceramics confirmed a cubic crystalline perovskite structure consistent with all previous studies [jcpds: 00-034-0411], with practically no impurity phase, and are shown in figure 1 [57–59]. The formation of single-phase Ba$_{0.6}$Sr$_{0.4}$Ti$_{1-x}$Al$_x$O$_3$ ($x = 0.00, 0.05, 0.10, 0.15, 0.20$) perovskites demonstrated doping with Sr$^{2+}$ ion at the A-site and doping with Al$^{3+}$ ion at the B-site might have almost incorporated into the perovskite structure of Barium Titanate. However, there are small amount impurities found for high Al doping concentration. The peaks with their corresponding Miller indices have been determined from the XRD patterns. The XRD peaks ($100$), ($101$), ($111$), ($200$), ($201$), ($211$), and ($220$), respectively, are well indexed to the crystal plane. Thus, the XRD studies revealed that all Al-doped compositions exhibit single peaks at 32° and 46°. To investigate the peak locations at various Al concentrations in more detail, the amplified XRD peaks ($2\theta = 32°–33°$), indicating the (110) plane, are presented in figure 1. The peak location for Al-doped compositions shifts from the lower diffraction angle toward the higher diffraction angle in this figure, indicating that the lattice parameter may have reduced with Al replacement because of the shrinking unit cell volumes. And the related results are listed in table 1. The Nelson–Riley extrapolation $F(\theta)$ was used to get each sample’s exact lattice constant values and calculated by the following equation, $F(\theta) = \frac{1}{2} \left[ \frac{\cos \theta}{\sin \theta} + \frac{\sin \theta}{\sin \theta} \right]$. Where $\theta$ indicates Bragg’s angle. This way, we can look for evidence of these findings. The computed values are also included in table 1. The lattice parameter ‘a’ for all diffracted planes is plotted versus the Nelson–Riley function $F(\theta)$, $x = 0–0.20$. The straight-line intercept obtained by the fit linear process was used to derive the lattice constant ‘a’ values [47]. The lattice parameter is inversely proportional to the ionic radius. It is expected that adding a divalent Al$^{3+}$ ion to the perovskite structure would result in a decrease in the ceramic samples’ lattice parameter ‘a.’ After studying the computed

![Figure 1](image-url)
lattice parameter values derived from the Nelson–Riley function and XRD data, it can be concluded that the lattice parameter values decline with the increase of Al contents up to 20% in both instances. Initially, the lattice constant for the undoped sample is 3.942 Å. After doping with Sr²⁺ and Al³⁺ contents, the lattice parameter of the samples falls to 3.941 Å, 3.937 Å, 3.930 Å, and 3.921 Å, respectively, as shown in figure 2. The cation distribution in the octahedral B-sites and tetrahedral A-sites has a significant effect on the electrical and magnetic properties of ceramics. Moreover, this lower lattice constant of the perovskite structure indicates that Sr²⁺ has replaced Ba²⁺ in the A-site since the ionic radius of Sr²⁺ (1.44 Å) is less than Ba²⁺ (1.61 Å). The B-site replacement of the Al³⁺ ion, which has a larger ionic radius of 1.84 Å than the Ti⁴⁺ (0.605 Å), might significantly influence the reduction of this lattice parameter. Finally, it was previously shown that when a more considerable Al³⁺ joins the crystal structure at the B-site, the unit cell expands, but the overall cubic symmetry remains intact [47].

3.1.1. Bulk and x-ray densities measurements
The bulk density and the x-ray density of the investigated ceramics with the varying degrees of doping are displayed in table 1 and shown in figure 3. The density of ceramics plays a significant role in determining their magnetic characteristics in ceramics. Also, it has been suggested that ceramics with higher densities may have higher permeability. The bulk density (B) of all compositions was determined using the mass-volume ratio, while the molecular weight and lattice constant were used to estimate the x-ray density (x). The difference in the undoped and substituted cations’ atomic weights [the atomic weight of Ti⁴⁺ (47.867 amu)] > the atomic weight of aluminum Al³⁺ (26.98 amu)] may have resulted in a drop in the ρB of Al-doped ceramics. In contrast, the development of the Al³⁺ concentration results in a considerable decrease in x-ray density. As the sum rule states that a mixture’s density is equal to the weighted sum of its component densities. The bulk densities are also less than the x-ray densities, as seen in figure 3(a).

![Figure 2. Variation of lattice parameter with Al content, x (mole) calculated from XRD data for Al-substituted Ba₀.₆Sr₀.₄Ti₁₋ₓAlₓO₃ composition (x = 0–0.20).](image-url)

Table 1. Structural components of ceramics, i.e., the lattice parameter, unit cell volume, Porosity and density of Al-doped Ba₀.₆Sr₀.₄Ti₁₋ₓAlₓO₃ composition (x = 0–0.20).

| Content, x (mole) | Samples | Molecular mass (gm) | Lattice parameter, c (Å) | Unit cell Volume, (a³) | Bulk density (g cm⁻³) | Theoretical (g cm⁻³) | Porosity P (%) |
|------------------|---------|---------------------|--------------------------|------------------------|-----------------------|-----------------------|-----------------|
| 0.00             | Ba₀.₆Sr₀.₄TiAlO₃ | 257.32              | 3.942                    | 61.256                 | 5.5                   | 6.905                 | 20.344          |
| 0.05             | Ba₀.₆Sr₀.₄Ti₀.₉₅Al₀.⁰₅O₃ | 255.87              | 3.941                    | 61.209                 | 5.33                  | 6.858                 | 22.281          |
| 0.10             | Ba₀.₆Sr₀.₄Ti₀.₉Al₀.₁O₃ | 254.43              | 3.937                    | 61.023                 | 5.25                  | 6.836                 | 23.212          |
| 0.15             | Ba₀.₆Sr₀.₄Ti₀.₈₅Al₀.₁₅O₃ | 252.98              | 3.930                    | 60.698                 | 5.16                  | 6.802                 | 24.067          |
| 0.20             | Ba₀.₆Sr₀.₄Ti₀.₈Al₀.₂O₃ | 251.53              | 3.921                    | 60.282                 | 5.08                  | 6.768                 | 24.940          |
3.1.2. Porosity

Two sources, such as intergranular and intragranular porosity can cause the total porosity. Consequently, the resultant porosity, P might be expressed as $P = P_{\text{inter}} + P_{\text{intra}}$. The intergranular porosity is significantly affected by grain size [60]. The porosity in the prepared samples, which may be generated and developed during sintering, exhibited an inverse pattern with bulk density (B), as the presence of pores in these samples may cause variation in bonding among the components of the compositions, resulting in a shift in the interatomic distance, which in turn increased the volume of the unit cell and lowered density [61, 62]. Figure 3(b) illustrates the impact of Al addition on the porosity and bulk density of the compositions. It is very evident that the bulk density of samples determined by the porosities decreases for x = 0.0–0.20. Therefore, there has been a considerable shift in the porosity values concerning the increment of Al concentration. The $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{Ti}_{1-x}\text{Al}_x\text{O}_3$ composition has the highest porosity of 24.94% and a minimum density of 6.768 g cm$^{-3}$. Besides, the porosity of the prepared samples may significantly affect how the crystallite size changes. Crystallite size may have increased because of the deterioration of crystallite’s grain boundaries. As a consequence of the addition of Al concentrations, our current study shows that porosity varies inversely with bulk density, which is similar to the previously published conclusion [61]. Table 1 summarizes the lattice parameter, density, porosity, and crystalline size, of several samples derived using the XRD data.

3.1.3. Williamson-Hall (W-H) Plot method

Crystallite size drops from 88 nm to 68 nm after initial doping in BST crystal and then grows to 76 nm with Al$^{3+}$ ion doping in BST crystals. The values of all ceramics were plotted using the Williamson-Hall (W-H) method, as illustrated in figure 4. And also listed in table 2. The plots exhibit an anisotropic character [61]. The crystallite sizes estimated by the W-H approaches were discovered to have similar behavior to those obtained using the Scherrer technique. Besides, the W-H method implies a larger crystallite size than the Scherrer method because the former considers microstrain, which induces peak widening [63].

3.2. Raman spectroscopy analysis

Raman spectroscopy is a valuable tool for studying ferroelectric materials due to the strong relation between lattice dynamics and ferroelectricity. ABO$_3$ perovskite materials having 15 degrees of freedom are divided into 1F$_{2u}$ + 4F$_{1u}$ above the cubic-tetragonal transition phase. In the cubic phase with Pm$n$3m or O$\text{h}$ point group, 1F$_{2u}$ and 3F$_{1u}$ relate to the optical branches, while the remaining F$_{1u}$ symmetrical mode refers to the acoustic branch.

Typically, ABO$_3$ perovskites have 12 optical modes in the paraelectric cubic phase, which change into the triply degenerated forms of O$\text{h}$ point group symmetry ($T_{\text{dab}} = 3F_{1u} + 1F_{2u}$). The F$_{2u}$ is considered silent mode because it is inactive in both Raman and infrared zone, while all F$_{1u}$ modes are infrared-active[64, 65]. The Raman spectra of prepared ceramics are presented in figure 5 at standard room temperature, ranging from 100 to 1200 cm$^{-1}$. The cubic phase has been detected in all of its notable modes, including broader E(TO1) ~ 230 cm$^{-1}$, A1(TO1) ~ 275 cm$^{-1}$, A1(TO2) ~ 375 cm$^{-1}$, E(TO2) ~ 450 cm$^{-1}$, A1(TO3) ~ 580 cm$^{-1}$, A$_0$(LO3)/E(LO) ~ 760 cm$^{-1}$ modes, respectively.

In brief, the cubic phase has been identified at room temperature from Raman spectra of all Al-doped and undoped ceramics [64, 66]. With increasing Al concentration, the stretching mode of A1(TO1) and A1(TO3) developed at about 275 cm$^{-1}$ and 580 cm$^{-1}$ moved to a slightly higher wavenumber direction with a low-

![Figure 3. (a) Variation of $\rho$, $\rho_B$ and $\rho_x$ with increasing Al content, x (mole) for Al-substituted $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{Ti}_{1-x}\text{Al}_x\text{O}_3$ composition ($x = 0–0.20$).](image-url)
Figure 4. Represents the W-H plots with increasing Al content, x (mole) for Al-substituted Ba$_{0.6}$Sr$_{0.4}$Ti$_{1-x}$Al$_x$O$_3$ composition ($x = 0–0.20$).

Figure 5. The Raman spectra with increasing Al content, x (mole) for Al-substituted Ba$_{0.6}$Sr$_{0.4}$Ti$_{1-x}$Al$_x$O$_3$ composition ($x = 0–0.20$).

Table 2. The crystalline size, lattice strain, and dislocation density of Al-doped BST are computed by the W–H method and Scherrer method.

| Composition (x) | Crystallite size | Dislocation density | Lattice strain |
|----------------|------------------|---------------------|---------------|
|                | D (nm) (scherrer) | D (nm) (W–H)        | $\delta$ (nm$^{-1}$) (scherrer) | $\delta$ (nm$^{-1}$) (W–H) | $\varepsilon$ (scherrer) | $\varepsilon$ (W–H) |
| 0.00           | 71.54            | 71.69               | 0.000 195     | 0.000 195     | 0.000 545     | 0.001 31      |
| 0.05           | 52.95            | 53.04               | 0.000 357     | 0.000 355     | 0.000 793     | 0.001 35      |
| 0.10           | 57.29            | 57.46               | 0.000 305     | 0.000 303     | 0.000 727     | 0.001 40      |
| 0.15           | 64.91            | 65.82               | 0.000 237     | 0.000 231     | 0.000 638     | 0.001 58      |
| 0.20           | 67.04            | 67.98               | 0.000 223     | 0.000 216     | 0.000 597     | 0.001 83      |
intensity border peak. Because these two Raman modes \( [A_1(TO1), A1(TO3)] \) are more symmetrical and much broader in the cubic phase \([67]\). From the figure, it was observed that the mode \( [E(TO2) \sim 450 \text{ cm}^{-1}] \) connected with the transition from tetragonal to cubic phase becomes broader or weaker with increasing \( Al \) concentration. It indicates only the cubic characteristics of the prepared samples. So, the tetragonal phase is ruled out from the prepared samples because of the disappearance of the sharper \( E(TO2) \sim 450 \text{ cm}^{-1} \) mode.

Since, in the cubic phase, the \( A1(LO3)/E(LO1) \) mode is exceedingly weak and broad, and it’s widely assumed that its sharp mode is only present in the tetragonal phase \([68, 69]\).

It was also noticeable that the intensity of \( A1(LO3)/E(LO1) \) mode observed at \( 760 \text{ cm}^{-1} \) decreases with increasing \( Al \) concentration. The modification of the Raman spectra may be attributed to the impact of lattice distortion, as \( Al^{3+} \) may substitute for \( Ti^{4+} \) at the \( B \)-site in the ceramics. The orthorhombic phase is also missing in the prepared ceramics because of the absence of Raman mode at the \( 485 \text{ cm}^{-1} \) wavenumber region \([70, 71]\).

Finally, it may be concluded from these Raman spectra that as \( Al \) increases, the characteristic peak intensities of all the bands undergo a strong decrease with a slight frequency range shift, suggesting that there is no phase change and that the prepared samples are shifting toward a more symmetrical cubic phase, pointing out the effect of \( Al \) on the BST structure. These observations are in excellent agreement with those indicated by the XRD investigation.

### 3.3. Fourier transforms infrared spectroscopy (FTIR) analysis

The FTIR spectra of aluminium doped Barium Strontium Titanate (Al-BSTO) synthesized at \( 1250 \) °C for 4 h are shown in figure 6. The presence of a sharp absorption band in the \( 400–4000 \text{ cm}^{-1} \) range suggests that the samples studied contain a significant quantity of \( H_2O \) and \( OH \) \([9, 72]\). In addition, the presence of a broad absorption band in the \( 400–4000 \text{ cm}^{-1} \) region indicates that the materials under investigation include a considerable amount of \( H_2O \) and \( OH \). In particular, the broad absorption band near \( 3400 \text{ cm}^{-1} \) region is related to the stretching vibration of the \( HO^- \) group with an intermolecular bond of hydrogen \([4, 73]\). It reveals the presence of \( HO^- \) group in the prepared samples for FTIR where KBr was used as a binder. As moisture was predicted since KBr is a sensitive substance.

Also, an absorption band observed in the low-frequency region of \( 450 \text{ cm}^{-1} \) to \( 850 \text{ cm}^{-1} \) was attributed to vibrations (vibrations of the \( TiO_6 \) octahedron) of the \( Ti-O \) isopropoxide. Therefore, the appearance of a sharp absorption peak at \( 560 \text{ cm}^{-1} \) confirms the vibration of the \( Ti-O \) bond \([74, 75]\). It is observed from the FTIR spectrum that the wavenumber of the sharp absorption peak shifts to the upper side with an increase in the concentration of \( Al \). When the concentration of \( Al \) increases from \( 0.00 \) to \( 0.05 \), the value of the wavenumber changes from around \( 520 \text{ cm}^{-1} \) to \( 540 \text{ cm}^{-1} \), \( 560 \text{ cm}^{-1} \) when \( Al = 0.10 \), \( 587 \text{ cm}^{-1} \) when \( Al = 0.15 \) and \( 597 \text{ cm}^{-1} \) when \( Al = 0.20 \). It denotes a reduction in cell size. The substitution of \( Al \) reduces the length of the \( Ti-O \) Bond.

### 3.4. Field dependent magnetization measurement by VSM

Magnetic hysteresis is important in determining the magnetic materials' potential uses. Figure 7 shows the \( M-H \) hysteresis curves for different \( Al^{3+} \) substituted \( Ba_{0.6}Sr_{0.4}Ti_{1-x}Al_xO_3 \) \( (x = 0.00–0.20) \) compositions. All ceramics exhibited fast magnetization growth at the beginning of the applied field and subsequently became saturated or
independent of the applied magnetic field. As a result, it’s evident that at room temperature, all of the samples are ferromagnetic \[47, 76\]. The three magnetic parameters of the compositions, including coercive field, anisotropy constant, and saturation magnetization, are computed from the hysteresis loops. And table 3 summarizes the results of M–H loop measurements for the samples. It is readily apparent from table 3 that the saturation magnetization for the composition with \(x = 0.05\) is 0.172 emu g\(^{-1}\), which is the maximum of all the ceramics. This finding was also supported by the increment of magnetic moment, permeability, and grain size. Extrinsic parameters like the ceramics’ microstructure and density also impact \(M_s\). Besides, it is well established that magnetization increases with density. The creation of domain walls becomes conceivable as grain size rises and magnetization improves because of magnetic field-induced domain wall displacement.

Consequently, the total value of magnetization is the sum of the contributions from all the elements that affect the composition. In the current investigation, magnetization of the sample increased for \(x = 0.05\) and subsequently reduced marginally for \(x = 0.1\) to 0.2. Reduced density increases pore count, which functions as pinning centers for electron spins, reducing the magnetization \[77\]. Furthermore, the reduction of saturation magnetization in these specific compositions is also due to the weakening permeability seen in figure 9 [the real component of permeability (\(\mu'\) versus frequency graph] [10% concentration to 20% concentration] \[78, 79\].

The existence of a diamagnetic component is typically related with a decrease in magnetization at high fields when \(x = 0.15\) and 0.2. The observed diamagnetism may have been caused by BaTiO\(_3\), which is diamagnetic by nature \[80\]. Besides, the substantial magnetic anisotropy may explain the observed anomaly in magnetization because it plays a significant role in the development of such behaviors, as shown by other compositions \[77\]. Also, tiny number of impurities such as aluminum or strontium oxides in composition may also shift the ferromagnetism at room temperature of materials which are not usually thought to be magnetic.

Notably, the coercive field quantifies the intensity of the magnetic field. Figure 8 shows how the saturation magnetization, Coercivity, and anisotropy constant change with different concentrations of Al. The Al\(^{3+}\) substitution has a noticeable effect on the Coercivity, \(H_c\). It is observed that Al substituted BSTAO composites have a higher coercivity than the parent material. The addition of Al content definitely improved anisotropy constant and Coercivity rather than saturation magnetization (Ms). According to magnetization theory, the three elements known as anisotropy constant, coercive field, and saturation magnetization are interconnected. In this research, Coercivity improved with dopants until \(x = 0.2\). The coercivity values may be influenced by

| Sample, x (mole) | Saturation magnetization, \(M_s\) (emu/g) | Coercivity, \(H_c\) (Oe) | Anisotropy constant \(K_a\) (emu/cm\(^3\)) |
|-----------------|----------------------------------------|----------------------|----------------------------------------|
| 0.00            | 0.048                                  | 395                  | 9.486                                  |
| 0.05            | 0.187                                  | 230                  | 21.446                                 |
| 0.10            | 0.174                                  | 460                  | 39.933                                 |
| 0.15            | 0.045                                  | 690                  | 15.525                                 |
| 0.20            | 0.035                                  | 940                  | 16.323                                 |

Figure 7. M–H hysteresis loops of Al–substituted Ba\(_{0.6}\)Sr\(_{0.4}\)Ti\(_{1-x}\)Al\(_x\)O\(_3\) composition (\(x = 0–0.20\)).

Table 3. The values of saturation magnetization, Coercivity, and anisotropy constant of Al-doped Ba\(_{0.6}\)Sr\(_{0.4}\)Ti\(_{1-x}\)Al\(_x\)O\(_3\) samples sintered at 1250 °C for 4 h.
parameters such as magnetocrystallinity, size distribution anisotropy, microstrain, and magnetic domain size.

Using the formula $K = \frac{HM_s}{2}$, the anisotropy constant (K) was derived. The values of $K_s$ were discovered to be raised up to the sample with all content, $x = 0.10$, as 39.9326 emu cm$^{-3}$ and subsequently dropped when the Al percentage was increased further because the produced samples tended to transition from 'soft' to 'hard' ferromagnets as the Al concentration increased. Increased anisotropy indicates that the composites are transforming into hard magnets. The composition $Ba_0.6Sr_0.4Ti_{1-x}Al_xO_3$ with an Al content ($x = 0.10$) had the highest $K_s$ value as well as a significant amount of coercive field ($H_c$). Magnetocrystalline anisotropy has a significant impact on ferromagnetic materials’ industrial applications. Materials having a high magnetic anisotropy often have a high coercivity, making them difficult to demagnetize. Permanent magnets are made from these hard ferromagnetic materials. Therefore, the sample with an Al concentration of $x = 0.10$ may be utilized to produce magnetic data storage devices.

3.5. Permeability measurements

Permeability is one of the essential factors for determining the magnetic property of perovskite (ABO$_3$) materials. The desirable magnetic attribute of perovskite is high permeability combined with negligible loss in the high-frequency range. High permeability is often found in materials with large grain sizes, low porosity, and high saturation magnetization.

The initial complex permeability may be expressed as $\mu'' = \mu_1' - i\mu_2''$ where, $\mu_1'$ and $i\mu_2''$ denote the real and imaginary components of the complex permeability, respectively. Since complex permeability is measured by magnetic domains’ dynamic responses to the applied field, it contains information about the domain walls’ inertial location, mutual interaction, and organization. At low frequencies, the permeability generates a great deal of information on the motion of the domain wall initially. Thus, the frequency-dependent information obtained from complicated permeability is essential for comprehending the process of dynamic characteristics as well as the efficacy of these materials in our everyday utilizable instruments that operate at high-frequency range. The change of the real component and the imaginary component of permeability ($\mu''$) with frequency for various concentrations of Al-doped Barium Strontium Titanate is shown in figure 3, respectively.

It was discovered that the values of $\mu''$ for each composition remained stable up to a specific frequency range before rapidly falling to minimal levels. The frequency at which $\mu''$ achieves its maximum value and suddenly falls is known as the resonance frequency, $f_r$. Resonance occurs when the applied frequency reaches the magnetic spins’ inherent frequency. Spinning and domain wall movements are the two most prominent ways of acquiring magnetization. Due to intra-granular pores and grain impurity, specific suppressed points were identified on the compositions’ surface, causing a drop of $\mu''$ at the high-frequency region. This effect causes an impediment to spin and domain wall motion. As a result, permeability drops dramatically, and loss rises. Notably, the oxygen vacancy concentration, which serves as a mechanical obstruction against the domain wall, may affect the domain wall’s motion. As a consequence of this change in oxygen vacancy concentration, the value of $\mu_1'$ fluctuates with the Al content. Aside from that, in the case of polycrystalline ceramics, the change of $\mu_1'$ with different samples is extremely difficult to define since $\mu_1'$ is affected by a number of parameters, including composition, stoichiometry, grain size, coercive field, crystal anisotropy, porosity, and impurity contents.

The influence of saturation magnetization, grain size, and anisotropy constant on permeability is described by the Globus equation, $\mu'' = M_s^2 D/K^{1/2}$, where, $K$ represents magnetocrystalline anisotropy constant, $M_s$ denotes saturation magnetization, and $D$ represents average grain size.

![Figure 8. (a) Variation of Saturation Magnetization, $M_s$ (b) Variation of Coercivity ($H_e$) and anisotropy constant ($K_a$) based on Al content, $x$ (mole) for Al-substituted $Ba_0.6Sr_0.4Ti_{1-x}Al_xO_3$ composition ($x = 0.0–0.20$).](Image 1)
Because of the $\text{Al}^{3+}$ substitution, the resonant frequency was changed towards a higher frequency area near 100 MHz, indicating the high-frequency stability of $\text{Al}^{3+}$ substituted compositions, as shown in figure 9. Here, the value of $\mu'_{i}$ is discovered as the $\text{Al}$ content increases, consistent with previous research \[85, 88\]. This constant permeability highlights that the measured frequency spectrum contains no resonance or structural relaxations. This also displays how long the ceramics can maintain their magnetic stability in changing applied fields. Because of their amazing steady permeability, the generated ceramics may be used throughout a large frequency range ($10^{-4}$ Hz–$10^{-8}$ Hz) \[79, 87\]. The result is consistent with Snoek’s rule, which states that initial permeability ($\mu'_{i}$) and resonance frequency ($f_{r}$) are connected as $\mu'_{i} f_{r} = \text{constant}$.

Figure 10(a) depicts the frequency-dependent permeability loss factor ($\tan \delta$). It indicates the magnetic system’s inefficiency. The value of $\tan \delta$ was observed to drop exponentially as the frequency increased. Due to the grain boundary, the low-frequency area has a high resistivity, which lowers eddy current losses. As a consequence, the loss in that area is significant. However, Grains contribute to low resistivity in the high-frequency range. This results in more excellent eddy current losses and lower magnetic losses. Besides, High frequency decreased the rate of domain wall motion, resulting in less loss \[47\]. The addition of a modest quantity of $\text{Al}$ content caused a minor change in the loss factor \[61\]. The composition with $\text{Al}$ content, $x = 0.10$, exhibits the most minor loss. It reduces in composites gradually as the $\text{Al}$ concentration rises, as indicated by the composites sum rule.

Figure 10(b) illustrates the variation in the RQF-factor permeability with the frequency of the synthesized perovskites. The frequency range over which the sample behaves as a hard magnetic material with minimal loss is determined by the permeability’s RQF-factor. It influences the magnetic material’s performance and quality. The perovskite composition with $\text{Al}$ content, $x = 0.10$ among the doped samples exhibited the highest RQF-factors, suggesting that it might be applied for high-frequency magnetic applications. Based on this data, the prepared ceramics have increased magnetic and dielectric properties, indicating that the compositions have higher application values. As a result, they may be used in headphones, computer hard drive magnets,
microphones, sensors, security systems, loudspeakers, generators like wave energy, wind turbines, turbo generators, magnetic suspensions, and motors like drills and washing machines.

3.6. Frequency-dependent dielectric constant measurement

The \( \varepsilon' \) reflects the material’s ability to accumulate charge compared to a vacuum by polarizing the medium in an applied electric field \[61\]. It may be thought of as the magnitude of the dynamical screening impact caused by a system’s charge excitations at a certain frequency.

Figure 11 shows the change of the real component \( (\varepsilon') \) of complex dielectric constant \( (\varepsilon^* = \varepsilon' - j\varepsilon'') \) for different \( Ba_{0.6}Sr_{0.4}Ti_{1-x}Al_xO_3 \) (where \( x = 0.00, 0.05, 0.01, 0.15, 0.20 \)) ceramics with a wider frequency range (5 kHz–5 MHz) at standard room temperature. This decreased frequency dispersion of \( \varepsilon' \) is the characteristic of ferroelectric and dielectric materials and may be explained by interfacial polarization postulated by Maxwell–Wagner \[89–91\], which agrees with Koop’s phenomenological hypothesis \[84\]. The compositional inhomogeneities produce interfacial defects, grain structure, impurities, and porosity, contributing to interfacial polarization. These inhomogeneities are introduced into the ceramics during pre-sintering and high-temperature sintering operations \[92\].

Polycrystalline ceramics, in general, are composed of thoroughly conducting grains partitioned by insulating grain boundaries in general. Charge carriers are displaced as a consequence of applying a field, and if the resistances are sufficiently high, charge carriers have a tendency to align themselves along grain boundaries \[93, 94\].

Additionally, the total dielectric constant depends on ionic, dipolar, electronic, and atomic polarization simultaneously \[95, 96\]. These polarizations, occurring more often at low frequencies, are frequency-dependent. As a consequence, the dielectric constant is increased at lower frequencies. Besides, the dipole relaxation concept states that at lower frequencies, dipoles can follow the frequency of the triggered electric field, which may explain the drop in dielectric constants with increasing frequency \[97–99\]. Therefore, at high frequencies, the net polarization decreases as all polarization mechanisms, save the electronic one, cease contributing. This is because only non-massive electrons are capable of keeping up with the rapidly changing applied field. Also, polarizability’s orientation and ionic sources reduced and eventually vanished at high frequencies region, which might be explained by molecule and ionic inertia \[100\]. This causes the frequency-independent dielectric constant to stabilize, referred to as the static dielectric constant. Moreover, the density and grain size (aforementioned) of ceramics significantly influence their dielectric constant. Thus, the undoped BST is more frequency sensitive than the doped BST.

The amplitude of \( \varepsilon' \) decreases with increasing Al concentration with compositional change due to high porosity \[92\]. Doping in perovskites, on the other hand, tends to form, tilt, or, change the BO\(_6\) octahedral orientation, which may limit polarizability \[101, 102\]. Another possible explanation for the lowering value of \( \varepsilon' \) with increasing Al content is the hopping electrons between Ti\(^{3+}\) and Ti\(^{4+}\), as well as a decrease in oxygen vacancies. This might clarify why doped BaTiO\(_3\) is frequency independent. Thus, across the examined frequency...
range, each doped sample demonstrates a nearly frequency-independent nature, enhancing the utility of the frequency band for multifunctional devices. This is a wonderful and valuable conclusion from our analyzed samples that may be used in a variety of applications.

The variation of the sample’s energy loss ($\tan \delta_E$) in the dielectric scheme is shown in figure 12(a). Because of the substantial friction between dipoles caused by the rapid alignment of dipoles in response to change in the applied electric field, the $\tan \delta_E$ of undoped BST is comparatively higher, supported by the sample’s greater $\varepsilon'$. The dielectric loss of the un-doped ceramics is found to be the lowest. Electronic conduction in perovskites is promoted by electron bouncing among identical ions that are freely scattered over crystallographically matching lattice locations, according to the hoping mechanism of Verway–de–Boer. At the lower frequencies domain, the electron hopping frequency between $\text{Ti}^{4+}$ and $\text{Ti}^{3+}$ ions are much higher compared to the applied ac field frequency, which maximizes the loss ($\tan \delta_E$). The loss, on the contrary, is reduced in the higher frequency region because the electron hopping frequency between $\text{Ti}^{4+}$ and $\text{Ti}^{3+}$ ions is incapable of chasing the applied field beyond a particular critical frequency. A considerable amount of energy is lost due to the restriction of alignments by other dipole effects. Moreover, ionic, electronic, and dipolar losses are present in all samples at low frequencies resulting in a substantial loss in that range. At higher frequencies, $\tan \delta_E$ has extremely tiny values, indicating that the dipoles having tiny effective masses (e.g., ferroelectric, electrons domains) assist the majority of the dielectric constant rather than Defects in charge (i.e., oxygen vacancies) having significant effective masses.

The sample with $x = 0.05$ may be a promising candidate for microwave device applications because of low loss tangent values in the higher frequency range. Furthermore, $\tan \delta_E$ is influenced by various parameters, including sintering temperature, stoichiometry, structural homogeneity, and compositional performance of the ceramics. Almost all doped ceramics have a frequency-independent response with decreased loss ($\tan \delta_E$), indicating that they might be used in microwave devices, oscillators, resonators, and magnetically adjustable filters.

Figure 12(b) depicts the RQF variation with frequency for prepared ceramics with $x$ ranging from 0.0 to 0.2. The RQF rises with increasing Al concentration up to $x = 0.05$ and subsequently falls from $x = 0.05$ to $x = 0.2$, dependent on real component and the loss factor of the dielectric constant. The ceramics with a higher dielectric constant may be utilized to manufacture semiconductors, gate dielectrics in MOS, primarily transistors, and electrically adjustable microwave devices. These samples may be used in high and low-frequency applications based on the RQF for dielectric analysis.

On the other hand, these ceramics appeal to spintronics devices and high-frequency microwave applications due to their negligible loss and high RQF of dielectric measurement. Besides that, they may also be utilized to manufacture magnetic cores for inductors and transformers.

3.7. Resistivity studies
Perovskite’s frequency-dependent AC resistivity is a metric for measuring its suitability for electronic applications. Figure 13(a) shows the resistivity variation from 1 kHz to 10 MHz. With increasing frequency, the resistivity value of all synthesized perovskites was observed to decrease. High AC resistivity values were observed...
in lower frequency regions, while higher frequencies increase the hopping rate, increasing the conduction process and decreasing resistivity \[107\].

The values of AC resistivity developed with increasing Al content, \(x\), due to the decrease in ion conductivity with increasing Al concentration, as shown in figure 13(b). Since high-resistive materials have a low eddy current loss, they’re essential for making incandescent bulbs, space heaters, calcination furnaces, and heating filaments, among other technical equipment and uses.

3.8. UV–Vis spectra and optical band gap analysis

UV analysis was performed on undoped and doped samples to determine the impact of \(\text{Al}^{3+}\) ions in BST. The diffuse reflectance spectra of pure and \(\text{Al}\)-doped BST ceramics at various concentrations are displayed in figure 14. Tauc’s relation \[43\] was used to compute the optical bandgap of each sample, as shown in figure 14: 

\[\alpha h\nu = A(h\nu - E_g)^n\]

where \(\alpha\) denotes the absorption coefficient, \(h\nu\) denotes the incident photon energy, \(n\) denotes a transition-specific parameter, \(A\) denotes the system-dependent parameter, and \(E_g\) denotes the optical band difference. The bandgap of all samples was estimated by graphing \((\alpha h\nu)^2\) Vs \(h\nu\), taking the greatest linear fit and projecting it toward the \(x\)-axis, as shown in inherited figure 14. It has been discovered that the bandgap energy of the samples doped with different \(\text{Al}^{3+}\) ions reduces from 3.23 eV to 3.02 eV, as shown in table 4. The bandgap of pure BST is higher than that of \(\text{Al}\) doped compositions. The difference in the bandgap of the prepared samples may be evaluated using the electronic states of HOMO (highest occupied molecular orbitals) and LUMO (lowest unoccupied molecular orbitals) as well as the electronegativity of dopant and host ions \[108, 109\]. The LUMO is around the \(d\) orbital of titanium (\(d\) (Ti)) and the \(s\) orbital of barium (Ba), whereas The HOMO is about the \(2p\) oxygen orbital (O) in a BST-based material. Thus, depending on the electronegativity of dopants, a new electronic state will be generated below or above the LUMO. According to the data, \(\text{Al}\) (1.61) has a higher electronegativity than \(\text{Ti}\) (1.54). Thus, \(\text{Ti}\) 3d orbitals have higher energy than \(\text{Al}\) 3s\(^2\) and 3p\(^1\) orbitals, causing the conduction band to move downward. As a result, it may be concluded that \(\text{Al}^{3+}\) doping reduces the band gap of the prepared samples. Furthermore, the band gap energy variations may also be confirmed by lattice parameters and the changes in electrical structure. The result is compatible with XRD research, which shows that adding \(\text{Al}\) dopant increases particle size \[110\]. Besides, the density, grain size, and oxygen deficit may all have a favorable influence on optical characteristics. All of the data above demonstrate that the optical characteristics of pure and doped BST powders can be readily regulated, indicating that the prepared samples are a potential choice for electro-optic purposes.

4. Conclusion

\(\text{Al}^{3+}\) substituted \(\text{Ba}_{0.6}\text{Sr}_{0.4}\text{Ti}_{1-x}\text{Al}_x\text{O}_3\) (\(x = 0, 0.05, 0.10, 0.15, \text{and} 0.20\)) ceramic samples were successfully synthesized using the conventional solid-state reaction technique. The structural, magnetic, electrical, and optical properties of the prepared samples were probed. The x-ray diffraction results revealed that all of the ceramics had a single-phase cubic perovskite structure, which was also validated by Raman spectroscopy. Both the XRD data and Nelson-Riley function showed that the lattice constant ‘\(a\)’ decreases from 3.942\(\text{Å}\) to 3.921\(\text{Å}\) with the increase of \(\text{Al}\) content. With the increase of \(\text{Al}\) content, the FTIR spectrum slightly blue shifts between
435 cm\(^{-1}\) to 534 cm\(^{-1}\) without constituting any secondary phase. The tauc plot demonstrates a slight reduction in the bandgap from 3.23 eV to 3.02 eV with the increase of Al content. The M–H curve obtained from VSM data.

**Table 4.** Band gap energy of Al-doped \(\text{Ba}_0.6\text{Sr}_0.4\text{Ti}_1-x\text{Al}_x\text{O}_3\) composition \((x = 0.0–0.20)\).

| Al content \(x\) (mole) | Band gap energy (eV) |
|--------------------------|----------------------|
| \(X = 0\)               | 3.23                 |
| \(X = 0.05\)             | 3.17                 |
| \(X = 0.1\)              | 3.12                 |
| \(X = 0.15\)             | 3.08                 |
| \(X = 0.2\)              | 3.02                 |

**Figure 14.** (a)–(e) UV–Vis transmittance spectra of different values of Al Content and (f) insert \([h\nu F(R)]^2\) versus photon energy, \(hv\) plots are used to determine the band gap energy of prepared ceramics.
demonstrates that all compositions have mild ferromagnetic characteristics at room temperature. It is found that the initial permeability is almost constant for all the samples up to the frequency range of about 75 MHz, and above this frequency range, it rapidly decreases which follows the Globus model. It is also revealed that at the high-frequency range, the prepared sample showed an excellent relative quality factor due to the low dielectric loss, particularly for the Al content of x = 0.05. The AC resistivity of the prepared sample increased with the increment of Al content, and the highest resistivity was found at x = 0.20, which exhibits the little eddy current loss characteristics of the sample. Although almost all of the compositions exhibit less magnetic & electrical behavior than the undoped sample, the samples with Al content (x = 0.05 & 0.10) showed better magnetic and electrical properties than the other compositions. Finally, it can be concluded that the compositions developed in this study may be employed as multiferroic materials in the industry to create prospective multifunctional devices with high-frequency performance.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

Completing interest

The authors declare no Completing Interest.

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