ABSTRACT: The structural, microstructural, and microwave dielectric properties of Ba$_{1-x}$Sr$_x$Ti$_4$O$_9$ (0.0 $\leq$ x $\leq$ 0.06) ceramics samples synthesized by a conventional route were investigated. These structural, microstructural, and dielectric properties were recorded using X-ray diffraction (XRD), scanning electron microscopy (SEM), and Fourier transform infrared (FTIR) and impedance analyzer spectroscopies. Ti–O octahedral distortion was observed due to Sr$^{2+}$ addition. The microwave dielectric properties were interrelated with various Sr$^{2+}$ concentrations. Excellent microwave dielectric properties, i.e., high relative permittivity ($\varepsilon_r = 71.50$) and low dielectric loss (tan $\delta = 0.0006$), were obtained.

INTRODUCTION

Microwave dielectric devices are used in advanced technological systems such as radar, satellite receiver modules, and mobile telephones. The dielectric material, which is used in telecommunication devices, is termed dielectric resonators (DRs). Dielectric resonators may be used to stabilize microwave oscillators and microwave filter frequencies. Barium titanate (BaTiO$_3$) and barium tetratitanate (BaTi$_4$O$_9$) are candidate materials for DRs in microwave telecommunication and satellite broadcasting. Microwave DRs provide important advantages in terms of stability, compactness, light weight, and comparatively low costs in the processing of high-frequency devices. The physical characteristics required for DRs are as follows.

(i) High relative permittivity ($\varepsilon_r$) to attain reduction of modules in the interpretation of $(1/\varepsilon_r^2)$, i.e., size dependence.

(ii) High quality factor ($Q \times f$) values to reduce tangent loss.

(iii) Small temperature coefficient of resonant frequency ($\tau_f$) for stabilization of resonant frequency.

BaTiO$_3$, BaTi$_4$O$_9$, and doped BaTi$_4$O$_9$ compounds meet these basic requirements for the application of DRs; for example, $\varepsilon_r = 39.11$, $Q \times f = 10,700$ GHz, and $\tau_f = +14.2$ ppm/°C. However, it is essential for the synthesized dielectric ceramics to have the required microwave dielectric properties. Thus, the processing of single-phase ceramics is necessary to investigate different characterizations. The mixed oxide route involves a high calcination temperature during the reaction of BaCO$_3$ and TiO$_2$ (raw materials), and secondary phases may also be formed during the calcination process. Additionally, the product may be contaminated with impurities from grinding media. Nagas et al. reported the effects of different additives on the phase and microwave dielectric properties of BaTiO$_3$ and BaTi$_4$O$_9$ ceramics. The dielectric and structural properties of BaTi$_4$O$_9$ with different dopants have been studied in the microwave-frequency range. The different types of additives, i.e., Sr in BaTi$_4$O$_9$, result in multiple phases including BaTi$_4$O$_9$, Ba$_2$Ti$_9$O$_{20}$, and TiO$_2$. Kolar et al. and Negas et al. determined the phases and investigated the microwave dielectric properties with a relatively lower frequency at 1 MHz. Several studies have been executed to improve the microwave dielectric properties of BaTi$_4$O$_9$ ceramics by doping different additives, i.e., Sr$^{2+}$, Ca$^{2+}$, Pb$^{2+}$, and Bi$^{3+}$ ions for Ba$^{2+}$ site-ion and Zr$^{4+}$ and Sn$^{4+}$ ions for Ti$^{4+}$ site-ion. Other synthesis routes such as sol gel and co-precipitation may also use to synthesize these products.

Barium tetratitanate (BaTi$_4$O$_9$) ceramics are one of the well-known dielectric materials and has been studied by numerous researchers for use in dielectric resonators, thermistors, and electro-optic devices. Due to its importance, in the present work, we studied the effect of Sr on phase, surface morphology,

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and the dielectric properties of BaTi₄O₉ ceramics using a controlled mixed oxide solid-state processing route to prepare (Ba₁₋ₓSrₓ)Ti₄O₉, 0.0 ≤ x ≤ 0.06. These products have been analyzed using X-ray diffraction (XRD), scanning electron microscopy (SEM), and Fourier transform infrared (FTIR) spectroscopy. The dielectric properties of products were measured using impedance spectroscopy.

## RESULTS AND DISCUSSION

### Phase Analysis

The XRD pattern of (Ba₁₋ₓSrₓ)Ti₄O₉, 0.0 ≤ x ≤ 0.06 sintered ceramics is shown in Figure 1. The XRD studies revealed the formation of the orthorhombic (Ammm) structured base composition of barium tetratitanate (BaTi₄O₉), which matches with ICDD/PDF card # 03-0070. It is suggested that Sr²⁺ is incorporated in the lattice of the base composition to partially replace Ba²⁺ ions. A shift of XRD peaks was detected toward higher Bragg-angle (2θ) values with increasing Sr²⁺ content in (Ba₁₋ₓSrₓ)Ti₄O₉. The shifting may be due to the substitution of relatively smaller cations of Sr²⁺ (Rₑ = 1.44 Å) for Ba²⁺ (Rₑ = 1.61 Å) following the Bragg diffraction law (2d Sin θ = mλ).²¹,²² A peak at 32.4° emerges with an increase in Sr²⁺, which is attributed to the transformation of the structure from orthorhombic (Ammm) at x = 0.0 to tetragonal (I₄/m) at x = 0.02, 0.04 and then to cubic (Pm₃m) at x = 0.06. The variation in lattice parameters with increasing Sr²⁺ content is attributed to the phase transition from orthorhombic to tetragonal and then to the cubic structure as listed in Table 1, while Table 2 represents the XRD data of the base composition (BaTi₄O₉).

The particle size and lattice strain of the Ba₁₋ₓSrₓTi₄O₉ (0.00 ≤ x ≤ 0.06) sample were determined using the Williamson–Hall (W–H) technique from the broadening of the XRD peaks.²³

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

(1)

The equation represents a straight line, where ε is the gradient (slope) of the line and kλ/D is the y-intercept. Consider the standard equation of a straight line,

\[
y = mx + c
\]

(2)

Now, we plot 4 sin θ on the x-axis and β cos θ on the y-axis. The value of the strain (ε_{W,H}) is given by the value of “m”, which represents the gradient (slope) of the line, and the crystallite size can be calculated from the y-intercept kλ/D.

Figure 2a–d shows Williamson–Hall (W–H) plots for Ba₁₋ₓSrₓTi₄O₉ (0.00 ≤ x ≤ 0.06) ceramics. The W–H is used for deconvoluting shapes (crystalline shapes) and strain that contributes to X-ray line broadening because Scherrer’s formula does not take into account the strain contribution. Therefore, the average crystallite size, dislocation density, and strain of W–H lie in the 2.944–11.612 Å, 0.74152 × 10⁻⁶–271.667 × 10⁻⁶, and 1.1949 × 10⁻³–22.871 × 10⁻³ ranges for Ba₁₋ₓSrₓTi₄O₉ (0.00 ≤ x ≤ 0.06) ceramics, respectively, as shown in Table 3.

Mathematically, the dislocation density (δ) was calculated using the equation²⁴

\[
\delta = \frac{1}{D^2}
\]

(3)

Dislocation strongly influences many other properties of materials. As the dopant element perfectly replaces the host ions in the crystal lattice, it influences the crystal structure and produces very small crystal defects that can be negligible.

The lattice strain (η) was calculated through the equation²⁵,²⁶

\[
\eta = \frac{\beta \cos \theta}{4}
\]

(4)

In Table 3, the deviation in the calculated lattice strain and crystallite sizes of all prepared Ba₁₋ₓSrₓTi₄O₉ (0.00 ≤ x ≤ 0.06) ceramics samples with compositions is shown. When the concentration of the dopant element is increased, the microstrain decreases due to the size of the dopant element being greater than the host ions, as shown in Table 3.

### Table 1. Structural Data of (Ba₁₋ₓSrₓ)Ti₄O₉, 0.0 ≤ x ≤ 0.06 Ceramics

| X    | structure | space group | a (Å)    | b (Å)    | c (Å)    |
|------|-----------|-------------|----------|----------|----------|
| 0.00 | orthorhombic | Ammm        | 6.29400  | 14.5324  | 3.79720  |
| 0.02 | tetragonal | I₄/m        | 10.1434  | 10.1434  | 2.96795  |
| 0.04 | tetragonal | I₄/m        | 10.1434  | 10.1434  | 2.96795  |
| 0.06 | cubic     | Pm₃m        | 3.89800  | 3.89800  | 3.89800  |

### Table 2. X-ray Diffraction Data for the Base Composition (BaTi₄O₉) at λ = 0.154 nm

| 2θ exp | 2θ calc | I exp | h | k | l | d exp | d calc |
|--------|---------|-------|---|---|---|-------|-------|
| 18.85  | 18.63   | 105.82| 1 | 2 | 0 | 4.70211| 4.75713|
| 22.45  | 23.14   | 140.18| 1 | 3 | 0 | 3.95557| 3.83916|
| 24.55  | 24.20   | 366.33| 0 | 1 | 1 | 3.62176| 3.67334|
| 28.20  | 28.34   | 163.72| 2 | 0 | 0 | 3.16072| 3.14543|
| 32.30  | 33.15   | 303.62| 1 | 3 | 1 | 2.76826| 2.69921|
| 35.30  | 36.08   | 184.69| 2 | 0 | 1 | 2.53956| 2.48643|
| 38.39  | 37.61   | 174.98| 2 | 1 | 1 | 2.34196| 2.38872|
| 42.55  | 42.92   | 192.57| 2 | 5 | 0 | 2.12212| 2.13312|
| 43.60  | 43.06   | 168.88| 3 | 0 | 0 | 2.07342| 2.09816|
| 45.65  | 45.99   | 206.25| 1 | 7 | 0 | 1.98496| 1.97107|
| 47.75  | 47.30   | 179.09| 2 | 6 | 0 | 1.90244| 1.91949|
| 49.60  | 49.59   | 156.41| 0 | 2 | 2 | 1.83573| 1.83607|
| 54.80  | 54.51   | 174.53| 0 | 4 | 2 | 1.67319| 1.68141|
| 56.85  | 56.56   | 199.23| 2 | 0 | 2 | 1.61762| 1.62522|

Figure 1. XRD pattern of (Ba₁₋ₓSrₓ)Ti₄O₉, 0.0 ≤ x ≤ 0.06 ceramics.
Microstructural Analysis. The SEM images of \((\text{Ba}_{1-x}\text{Sr}_x\text{Ti}_4\text{O}_9, 0.0 \leq x \leq 0.06)\) ceramics sintered at 1300 °C in air for 2 h, polished, and thermally etched are shown in Figure 3. The SEM images indicated a dense microstructure with no obvious pores and grains, exhibiting elongated platelike morphologies for the base composition \((x = 0.00)\), which is consistent with previous reports for the orthorhombic-structured BaTi4O9.27,28 The grain morphologies were observed to change from elongated to rectangular with an increase in the Sr2+ content. The grain size for \((x = 0.00)\) is about 10 \(\times\) 1 \(\mu\)m and decreases with increasing Sr2+ content. The variation of relative densities \((\rho_r)\) with increasing Sr2+ content is shown in Table 4. The maximum theoretical density achieved is 4.94 g/cm\(^3\) as listed in Table 3. The increase in density may affect the value of the dielectric constant.11

FTIR Spectroscopy. The FTIR spectra of \((\text{Ba}_{1-x}\text{Sr}_x\text{Ti}_4\text{O}_9, 0.0 \leq x \leq 0.06)\) ceramics have been studied as shown in Figure 4. Very strong absorption peaks appear near 800 and 1400 cm\(^{-1}\) at \(x = 0.00\), while minor peaks are also observed at \(x > 0.00\) near 1600 cm\(^{-1}\). These peaks revealed the Ti–O octahedral vibrations according to previous studies on titanates.29 Due to the addition of Sr2+, the concentration in the solid solution of BaT4O9 ceramics is been shifted to higher wavenumbers. Sun et al. reported that only one oxygen vacancy can be used to replace Ba2+ ion, while the remaining three oxygen vacancies were used to replace the produced Ti4+ ion using respective additives.30 Therefore, Ti–O octahedra are easily distorted or damaged in this way. Some vibrational modes were observed in the FTIR spectrum. Therefore, relative studies of the FTIR spectrum further support the development of redispersibility of polycrystalline BaT4O9 ceramic dielectrics.

Microwave Dielectric Properties. The microwave dielectric properties of \((\text{Ba}_{1-x}\text{Sr}_x\text{Ti}_4\text{O}_9, 0.0 \leq x \leq 0.06)\) ceramics have also been investigated, as shown in Figure 5. The dielectric constant \((\varepsilon_r)\) varies from 21.9 to 71.50, while the variation in dielectric loss (tan \(\delta\)) with Sr2+ is shown in Table 4. The maximum value of tan \(\delta\) (0.0006) was observed. The frequency-dependent quality factor \((Q)\) is a dimensionless physical quantity, and quantitatively, it is expressed in terms of \(Q \times f\).31–35

The variations of tan \(\delta\) with frequency \((f)\) for various Sr2+ contents in \((\text{Ba}_{1-x}\text{Sr}_x\text{Ti}_4\text{O}_9, 0.0 \leq x \leq 0.06)\) are shown in Figure 6. The orientation polarization decreases with increasing frequency and results in an increase in dielectric loss, which may be attributed to the time lag between flipping dipoles and the applied electric field.13,6–38

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**Figure 2.** Williamson–Hall plot of Ba1-xSr_xTi4O9 with Sr content (a) \(X = 0.00\), (b) \(X = 0.02\), (c) \(X = 0.04\), and (d) \(X = 0.06\).

**Table 3.** Williamson–Hall (W–H) Calculated Crystallite Size \((D_{W-H})\), Dislocation Density \((\delta_{W-H})\), and Strain \((\eta_{W-H})\) for \((\text{Ba}_{1-x}\text{Sr}_x\text{Ti}_4\text{O}_9, 0.0 \leq x \leq 0.06)\) Ba1-xSr_xTi4O9.

| Composition | \(D_{W-H}\) (nm) | \(\delta_{W-H}\) \((\times 10^{-6} \text{ nm}^{-2})\) | \(\eta_{W-H}\) \((\times 10^{-3})\) |
|-------------|------------------|-----------------|----------------|
| 0.00        | 0.29449          | 11.5301         | 4.7118         |
| 0.02        | 0.06067          | 271.667         | 22.871         |
| 0.04        | 0.13191          | 57.4710         | 10.519         |
| 0.06        | 1.16128          | 0.74152         | 1.1949         |

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**Table 3.** Williamson–Hall (W–H) Calculated Crystallite Size \((D_{W-H})\), Dislocation Density \((\delta_{W-H})\), and Strain \((\eta_{W-H})\) for \((\text{Ba}_{1-x}\text{Sr}_x\text{Ti}_4\text{O}_9, 0.0 \leq x \leq 0.06)\) Ba1-xSr_xTi4O9.
dielectric material is usually characterized by high relative permittivity and a low tangent loss. The theoretical justification is very important for this case in which ionic crystals with an optical mode of vibrations resonate at a frequency of \((10^{13}\, \text{Hz})\). In the frequency range from approximately \(10^9\) to \(10^{11}\) Hz, the dielectric dispersion theory shows the contribution to polarization from the ionic displacement to be nearly constant and the loss to increase with frequency.\(^{39}\)

### CONCLUSIONS

The structural, microstructural, and microwave dielectric properties of \((\text{Ba}_{1-x}\text{Sr}_x)\text{Ti}_4\text{O}_9\), \(0.0 \leq x \leq 0.06\) sintered ceramics were investigated via a solid-state route. It is found that the dielectric constant \((\varepsilon_r)\) and dielectric loss \((\tan \delta)\) values improved with Sr\(^{2+}\) content. The \((\text{Ba}_{0.98}\text{Sr}_{0.02})\text{Ti}_4\text{O}_9\)

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**Table 4. Density Parameters and Dielectric Properties of \((\text{Ba}_{1-x}\text{Sr}_x)\text{Ti}_4\text{O}_9, 0.0 \leq x \leq 0.06\) Ceramics**\(^a\)

| \(x\) | \(\rho_a\) (g/cm\(^3\)) | \(\rho_t\) (g/cm\(^3\)) | \(\rho_r\) (%) | \(\tan \delta\) | \(\varepsilon_r\) |
|------|----------------|----------------|-------------|-------------|-------------|
| 0.00 | 4.26           | 4.402          | 96.77       | 0.0008      | 21.93       |
| 0.02 | 4.40           | 4.840          | 90.90       | 0.0006      | 47.84       |
| 0.04 | 4.40           | 4.940          | 89.07       | 0.0043      | 71.50       |
| 0.06 | 4.07           | 4.540          | 89.64       | 0.0044      | 52.50       |

\(^a\)Note: \(\rho_a\) = apparent density, \(\rho_t\) = theoretical density, and \(\rho_r\) = relative density.
ceramics was found to have the best $\varepsilon_r$ and $\tan \delta$ values. The microwave dielectric properties of (Ba$_{0.98}$Sr$_{0.02}$)Ti$_4$O$_9$ ceramics intensely depend upon density. Outstanding microwave dielectric properties of $\varepsilon_r \sim 47.84$ and $\tan \delta \sim 0.0006$ were obtained for (Ba$_{0.98}$Sr$_{0.02}$)Ti$_4$O$_9$ ceramics sintered at 1300 °C for 2 h. We obtained excellent microwave dielectric properties in this study for the application of microwave wireless communication systems.

## EXPERIMENTAL PROCEDURE

BaCO$_3$ (purity 99.0%, Chemdad, China), SrCO$_3$ (purity 99.5%, ABCSO, U.K.), and TiO$_2$ (purity 99.9%, Sigma) were chosen as raw starting materials to prepare (Ba$_{1-x}$Sr$_x$)Ti$_4$O$_9$, 0.0 $\leq x \leq$ 0.06 ceramics devices. The raw materials, i.e., BaCO$_3$, SrCO$_3$, and TiO$_2$, were thoroughly mixed according to the (Ba$_{1-x}$Sr$_x$)Ti$_4$O$_9$, 0 $\leq x \leq$ 0.06 stoichiometric ratios, where the mole ratio of A site-ion and B site-ion was 1:4. Distilled water was added to the weighed raw material powder in a polyethylene jar container along with 5 mm diameter zirconia balls and then milled by horizontal ball milling for 24 h. The mixture powders were dried at 100 °C for 24 h in a air atmosphere, and after drying, the reactant mixture was loaded in an alumina crucible and calcined at 1000 °C for 3 h in air at 10 °C/min in a heating/cooling rate. After calcination, the product powder was ground and then pressed into green body discs (5 mm thickness and 10 mm diameter) under a pressure of 80 MPa using a manual pellet press (CARVER). The pellet samples were sintered at 1300 °C for 2 h in air with a heating/cooling rate of 10 °C/min. The crystalline phases of the calcined (Ba$_{1-x}$Sr$_x$)Ti$_4$O$_9$, 0.0 $\leq x \leq$ 0.06 ceramics samples were identified using X-ray diffraction (XRD) (JDX-3532, JEOL, Japan) with a Cu Kα (λ = 0.15406 nm) radiation source operated at 40 mA and 40 kV in a wide range of Bragg’s angle 2θ (20° $\leq$ 2θ $\leq$ 80°) at a scanning rate of 2°/min. The surface morphology information was obtained using SEM (JSM-5910, JEOL Japan), while the microwave dielectric properties of the sintered samples were measured at microwave frequencies using impedance spectroscopy (Agilent 4287A).

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