Effects of Strontium on the Structural, Optical, and Microwave Dielectric Properties of Ba$_2$Ti$_9$O$_{20}$ Ceramics Synthesized by a Mixed Oxide Route

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ABSTRACT: Solid solutions of Sr-doped barium nonatitanate (Ba$_2$Ti$_9$O$_{20}$) ceramics were synthesized by a mixed oxide route, studied their structural, microstructural, optical, and microwave dielectric properties were studied. X-ray diffraction (XRD) has been used to reveal the structure and crystallite size of doped Ba$_2$Ti$_9$O$_{20}$ ceramics. Rietveld refinements of XRD patterns revealed that all of the ceramics have a tetragonal structure with space group $I4/m$. The surface morphologies of all the samples were characterized by using scanning electron microscopy (SEM). Fourier transform infrared (FT-IR) studies gave the O−H and Ti−O modes of vibrations. The stretching modes of Ti−O and O−H were noted at 1400, 2960, 3700, and near to 440 cm$^{-1}$, respectively. The band gap energy decreases with increasing Sr$^{2+}$ contents, and a high value (2.12 eV) was observed in the base sample. The microwave dielectric properties of the ceramic samples were studied at different frequencies ranging from 1 to 2 GHz by using impedance spectroscopy. The obtained results showed the suitability of these samples for microwave dielectric resonator (antenna) applications.

INTRODUCTION

Due to their outstanding structural and dielectrical properties, barium-based titanate ceramics such as BaTi$_4$O$_9$, BaTi$_3$O$_{11}$, and Ba$_2$Ti$_9$O$_{20}$ have been traditionally observed to be emerging materials. The Ba$_2$Ti$_9$O$_{20}$ (barium nonatitanate) ceramic has attracted interest due to the importance of its many applications in the field of modern telecom industries: i.e. radio stations (software base), satellites for monitoring environmental parameters, and GPS (global positioning systems). In addition, this ceramic has good-quality microwave dielectric resonator components for high-speed communications devices, including high-capacity data storage devices, rechargeable batteries, and dielectric resonators. To make improvements to the barium-based compound, to give it excellent dielectric qualities at a reasonable cost, most enterprises and research have provided microlevel oscillators and low-frequency microwave analyzers or filters for these purposes.$^{12}$ Ba$_2$Ti$_9$O$_{20}$, BaTi$_4$O$_9$, and BaTi$_3$O$_{11}$ exhibit high values of relative permittivity and a low dielectric loss in the radio frequency range.$^3$ Furthermore, a complex perovskite $A$$_2$B$_2$O$_{12}$ type barium nonatitanate (Ba$_2$Ti$_9$O$_{20}$) has been studied where the A and B sites generally represent cations while the O site represents an anion.$^4$−$^7$ Meanwhile, doping divalent metals (Ca$^{2+}$, Sr$^{2+}$, Sn$^{2+}$) in Ba$_2$Ti$_9$O$_{20}$ modified the microwave properties, i.e. the dielectric constant ($\varepsilon_r$) of the ceramic pellets, the quality factor ($Q_f$), and the temperature coefficient at the resonant frequency ($\tau_f$), which are very necessary for making microwave wireless communication devices. In addition, the Sr$^{2+}$ concentrations modified the microwave dielectric properties of the base ceramic material. Ba$_2$Ti$_9$O$_{20}$ has good properties, i.e. a high value of the relative permittivity ($\varepsilon_r = 38.4$), a good temperature coefficient of the resonant frequency ($\tau_f = +20$ ppm °C$^{-1}$), a high quality factor ($Q_f = 3300$ GHz), and low dielectric loss (tan $\sigma = 0.0056$), and solid solutions of doped concentrations of Ca, Sr, Zr, or Sn in Ba$_2$Ti$_9$O$_{20}$ ceramics are very suitable for making good dielectric resonator antennas.$^8$−$^{11}$ For Ba$_2$Ti$_9$O$_{20}$ sintered ceramics, it is necessary to get maximum values of the quality factor ($Q_fX_f$) and low values of the temperature coefficient at the resonant frequency ($\tau_f$). BaSnO$_3$ and BaWO$_4$ have negative $\tau_f$ values, which when they are added to the aforementioned dopants give
positive \( \tau \) values to give a nearly zero \( \tau \) value, but the adjustment of \( \tau \) and \( Q \) is very difficult.\textsuperscript{12–14} Many researchers have studied the effects of different dopants on the structural and microwave dielectric properties of Ba\(_4\)Ti\(_4\)O\(_{20}\) sintered ceramics.\textsuperscript{15}

The objective of the present work is to study the effects of strontium carbonate on the microstructure development, crystal structure, and dielectric properties of solid solutions of \((\text{Ba}_{1-x}\text{Sr}_x)\text{Ti}_4\text{O}_{20}\) (0.00 \( \leq x \leq 0.06\)) ceramics. The results of the work show that the influence of Sr\(^{2+}\) contents modifies the phase, microstructure, stretching vibration, and microwave properties (i.e., relative permittivity and quality factor) of the Ba\(_4\)Ti\(_4\)O\(_{20}\) materials.

## RESULTS AND DISCUSSION

**Phase Analysis.** The XRD profile of \((\text{Ba}_{1-x}\text{Sr}_x)\text{Ti}_4\text{O}_{20}\) (0.00 \( \leq x \leq 0.06\)) ceramics with various Sr contents are shown in Figure 1a,b. The tetragonal structure of \(I4/m\) (space group) basic contents of Ba\(_4\)Ti\(_4\)O\(_{20}\) ceramics matched with that of JCPD card number 01-080-0920, which represents the Sr carbonate compound with high-intensity, sharp diffraction peaks (Figure 1). This might be ascribed to microstrain, inhomogeneity in the samples, or the replacement of the Ba\(^{2+}\) \((R_{\text{Ba}} = 1.42 \text{ Å})\) cation by the comparatively larger radius of the Sr\(^{2+}\) \((R_{\text{Sr}} = 1.44 \text{ Å})\) cation.\textsuperscript{16}

The effect of Sr concentrations on the crystallite size when Sr was integrated in \((\text{Ba}_{1-x}\text{Sr}_x)\text{Ti}_4\text{O}_{20}\) structure was evaluated using Scherrer’s formula\textsuperscript{17}

\[
D = \frac{K\lambda}{\beta \cos \theta}
\]

where \( \lambda \) represents the wavelength of the X-rays used (\( \lambda = 1.5406 \text{ Å} \)), while \( \beta \) gives full width at half-maximum (fwhm) in radians, \( D \) is the average crystallite size, and \( \theta \) is half of the angle between the incident and reflected X-ray beams.

The calculated values of the lattice strain are given in Table 1. The lattice strain increases with an increase in strontium content up to \( x = 0.04 \); above this value the lattice strain decreases. The increase of lattice strain decreases the crystallite size up to \( x = 0.04 \), and at higher Sr doping levels, the ionic radius affects the crystallite size because the ionic radius of strontium is greater than that of barium.

The dislocation density can be determined with\textsuperscript{19}

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

where \( \delta \) is the dislocation density while \( D \) is the crystallite size.

The crystalline phase (tetragonal structure) of the synthesized compound with high-intensity, sharp diffraction peaks (Figure 1) may be due to an increase in the Sr contents and the phase formation as well. The relative density also affected the structural properties of the ceramic samples. The relative density can be calculated from the experimental and theoretical densities of sample as shown in Table 1. The theoretical density \( \left( \rho_{th} \right) \) of a sample can be calculated by using the formula

\[
\rho_{th} = \frac{ZM}{N_A}
\]

where \( Z \) represents the number of atoms per unit cell while \( M \) and \( N_A \) are the molecular weight and Avogadro’s number, respectively. Experimental density was measured by using the Archimedes principle.\textsuperscript{20}

A Rietveld refinement was performed to confirm the exact the phase and lattice parameters of these ceramics using FullProf software. The refinement was carried out using the \(I4/m\) space group. The observed, calculated, and difference X-ray diffraction patterns for \((\text{Ba}_{1-x}\text{Sr}_x)\text{Ti}_4\text{O}_{20}\) at \( x = 0.0, 0.02, 0.04, 0.06 \) are displayed in Figure 2a–d, respectively.

**Microstructural Analysis.** The SEM images of gold-coated and thermally etched samples of \((\text{Ba}_{1-x}\text{Sr}_x)\text{Ti}_4\text{O}_{20}\) (0.00 \( \leq x \leq 0.06\)) sintered pellets are shown in Figure 3. The grain size variations and surface morphologies of the samples have been

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**Table 1. Properties of \((\text{Ba}_{1-x}\text{Sr}_x)\text{Ti}_4\text{O}_{20}\) (0.00 \( \leq x \leq 0.06\)) Ceramics**

| content \((x)\) | structure | space group | \(\rho_{\text{exp}}\) \((\text{g cm}^{-3})\) | \(\rho_{th}\) \((\text{g cm}^{-3})\) | \(\rho_{r}\) \((\%\)) | \(D\) \((\text{nm})\) | \(\delta\) \((\text{nm}^{-2})\) | \(\eta\) \((\times 10^{-5})\) |
|---|---|---|---|---|---|---|---|---|
| 0.00 | tetragonal | \(I4/m\) | 4.54 | 4.72 | 95.98 | 7.0513 | 0.02011 | 0.49159 |
| 0.02 | tetragonal | \(I4/m\) | 4.30 | 4.64 | 94.82 | 2.4559 | 0.16579 | 1.41143 |
| 0.04 | tetragonal | \(I4/m\) | 4.27 | 4.62 | 92.42 | 1.3979 | 0.51176 | 2.47973 |
| 0.06 | tetragonal | \(I4/m\) | 4.18 | 4.56 | 91.66 | 2.2393 | 0.19943 | 1.54798 |

*Definitions: \(\rho_{\text{exp}}\) experimental density; \(\rho_{th}\) theoretical density; \(\rho_{r}\) relative density; \(D\), average crystallite size; \(\delta\), dislocation density; \(\eta\), lattice strain.*
investigated. The SEM images of the sintered Ba$_2$Ti$_9$O$_{20}$ ceramic without and with glass or Mn additions at various sintering temperatures have been revealed by many researchers. The pure surfaces of Ba$_2$Ti$_9$O$_{20}$ ceramics was observed to have pores and no grain growth. A decrease in porosity and the production of new grains was observed in Ba$_2$Ti$_9$O$_{20}$ ceramics with glass dopants sintered at 1020 °C. The effects of glass additions to Ba$_2$Ti$_9$O$_{20}$ ceramics were modified grain growth and a low sintering temperature. SEM images showed the surface morphology and porosity of all prepared pellets. The porosity and grain size increases with increasing Sr concentrations in Ba$_2$Ti$_9$O$_{20}$ sintered ceramics (Figure 3). These factors may affect the phase and microwave dielectric properties. For this purpose several researchers adopted and controlled the synthesis parameters during the preparation of ceramic dielectrics, which modified the microstructural dielectric properties. Fourier Transform Infrared (FTIR) Spectroscopy. The FTIR spectra of (Ba$_{1-x}$Sr$_x$)$_2$Ti$_9$O$_{20}$ (0.00 ≤ x ≤ 0.06) sintered ceramics are shown in Figure 4. To study the procedure of the chemical reaction for the preparation of a ceramic material, FTIR spectroscopy plays a vital role in analyzing the stretching and vibrational modes. The O−H stretching mode was reported at different wavenumbers: i.e. 1400, 2960, and 37000 cm$^{-1}$. This mode may occur due to the presence of humidity in the characterized samples. Only one asymmetric mode (near the wavenumber 1050 cm$^{-1}$) has been reported, which showed that the carboxylate group bonded along with Ba entities. In a given spectrum, a normal stretching vibrational mode (Ti−O) appears near the wavenumber 440 cm$^{-1}$.

Figure 2. Rietveld refined XRD pattern of (Ba$_{1-x}$Sr$_x$)$_2$Ti$_9$O$_{20}$ for (a) x = 0.0, (b) x = 0.02, (c) x = 0.04, and (d) x = 0.06 ceramics.

Figure 3. SEM images of gold-coated (Ba$_{1-x}$Sr$_x$)$_2$Ti$_9$O$_{20}$ sintered ceramics: (a) x = 0.00; (b) x = 0.02; (c) x = 0.04; (d) x = 0.06.
Optical Studies. Figure 5 shows the UV–vis optical absorption spectra of (Ba$_{1-x}$Sr$_x$)$_2$Ti$_9$O$_{20}$ ($0.00 \leq x \leq 0.06$) ceramics. It can be seen that the base sample exhibits an optical response at wavelengths shorter than 426 nm. This shows that the base sample is active under UV light and a narrow region of visible light. The light absorption edges in the UV optical absorption spectra of doped samples were extended to the visible light region (450–600 nm). The optical absorption band edge can be expressed by the Tauc equation (eq 5)

$$\alpha = \frac{A(h\nu - E_g)^{1/2}}{h\nu}$$

where $E_g$ is the band-gap energy and $A$ is the proportionality constant while $h\nu$ is the energy of a photon. By using a Tauc plot, the optical absorptions and band-gap energies ($E_g$) for all of the samples were calculated, as shown in the Figure 4. The natural transitions (electronic) of the samples have been studied by this method.

Many researchers have reported that the Ba$_2$Ti$_9$O$_{20}$ sample is translucent for white light. It is very important to note that Ba$_2$Ti$_9$O$_{20}$ ceramics are translucent in the visible spectrum.$^{25}$ The structural and transitional bands have been analyzed with the help of the photon energy.$^{26}$ To obtain an optical band gap energy, the electron should undergo inner-shell transitions. This band depends upon the absorption coefficient ($\alpha$), which has been calculated by using eq 5.

AC Conductivity. AC conductivity measurements were made at room temperature for all the samples in the frequency range 1–2 GHz using an impedance analyzer. A frequency-dependent AC electrical conductivity study was carried out. The

Figure 4. FTIR spectra of (Ba$_{1-x}$Sr$_x$)$_2$Ti$_9$O$_{20}$ ($0.00 \leq x \leq 0.06$) ceramics.

Figure 5. Absorption spectra and plots of the band gap energies of (Ba$_{1-x}$Sr$_x$)$_2$Ti$_9$O$_{20}$ ceramics: (a) $x = 0.0$; (b) $x = 0.02$; (c) $x = 0.04$; (d) $x = 0.06$. 
The frequency-dependent AC conductivity of the \((\text{Ba}_{1-x}\text{Sr}_x)_2\text{Ti}_9\text{O}_{20}\) ceramics are shown in Figure 6. The AC conductivity (frequency dependent) was determined by eq 6:

\[
\sigma_{\text{AC}} = \epsilon_0 \epsilon_r \omega^2 \tan \delta
\]

where \(\epsilon_r\) is the dielectric constant, \(\epsilon_0\) is the permittivity of free space \((8.85 \times 10^{-14} \text{ F/cm})\), \(\tan \delta\) is the loss tangent, and \(\omega^2\) is the angular frequency \((\text{i.e., } \omega = 2\pi f)\). The AC conductivity increases slowly at low frequency but abruptly increases at high frequency. These changes in AC conductivity may occur due to the hopping charge carriers and structure of the grains as well.\(^{28,29}\) It is observed that the conductivity \(\sigma_{\text{AC}}\) varies with an increase in frequency. The trapping and fluctuations of conductivity may be due to the increase in temperature of the ceramic samples from the thermal activation energy. Due to the thermal activation energy, the movement of charge carriers increases inside the dielectric resonators.

**Dielectric Properties.** In the current research work, the dielectric properties, i.e. the relative permittivity and quality factor, has been measured in the frequency range 1–2 GHz. The dielectric constant value of the \((\text{Ba}_{1-x}\text{Sr}_x)_2\text{Ti}_9\text{O}_{20}\) sintered ceramics changes with an increase in the Sr concentrations. The obtained dielectric constant values for \(\text{Ba}_2\text{Ti}_9\text{O}_{20}\), \((\text{Ba}_{0.98}\text{Sr}_{0.02})_2\text{Ti}_9\text{O}_{20}\), \((\text{Ba}_{0.96}\text{Sr}_{0.04})_2\text{Ti}_9\text{O}_{20}\), and \((\text{Ba}_{0.94}\text{Sr}_{0.06})_2\text{Ti}_9\text{O}_{20}\) were 28.87, 31.21, 32.78, and 36.93, respectively, at a frequency of 1 GHz. This shows that a small increase in the Sr concentration increased the relative permittivity values. Figure 7 shows that the value of the dielectric constant decreases with an increase in the operating frequency. The value of dielectric constant can be explained easily according to the relative dipole moments and lattice structures of the \((\text{Ba}_{1-x}\text{Sr}_x)_2\text{Ti}_9\text{O}_{20}\) samples. In the cubical structure of \((\text{Ba}_{1-x}\text{Sr}_x)_2\text{Ti}_9\text{O}_{20}\) ceramics, the Ti atom coordinated octahedrally with 6 oxygen atoms. To obtain a permanent dipole moment, Ti atoms took centrosymmetric positions on the \(c\) axis, which enhanced the values of the relative permittivities. It was concluded that tetragonality depends upon the \(c/a\) ratio and the higher this ratio is, the relative permittivity maximum will be.\(^{30,31}\)

![Figure 6. Variation of AC conductivity with frequency of the \((\text{Ba}_{1-x}\text{Sr}_x)_2\text{Ti}_9\text{O}_{20}\) ceramics.](image)

The quality factor and relative permittivity both follow same the tendency with a variation in frequency: i.e. these values are the minimum and the maximum in high- and low-frequency regions, respectively. The quality factor variation along with frequency of \((\text{Ba}_{1-x}\text{Sr}_x)_2\text{Ti}_9\text{O}_{20}\) ceramics is shown in Figure 8. In the low-frequency region, the values of the quality factor strongly depend upon the frequency while in the high frequency region there appears to be a saturation phase. The quality factor of the base sample \((x = 0.00)\) decreases with Sr concentration and nearly vanishes at the content \(x = 0.02\); due to space charge polarization the values of the quality factor may be affected. The quality factor is the ratio of energy stored per cycle to the energy dissipated per cycle of the operating AC source.\(^{12}\) It was observed that the quality factor values decrease with an increase in the Sr concentrations.

![Figure 7. Frequency-dependent relative permittivity of \((\text{Ba}_{1-x}\text{Sr}_x)_2\text{Ti}_9\text{O}_{20}\) \((0.00 \leq x \leq 0.06)\) ceramics.](image)

![Figure 8. Variation of quality factors of \((\text{Ba}_{1-x}\text{Sr}_x)_2\text{Ti}_9\text{O}_{20}\) \((0.00 \leq x \leq 0.06)\) ceramics with frequency.](image)
(\(Ba_{1-x}Sr_x\))\(_2\)Ti\(_3\)O\(_9\) (0.00 \(\leq x \leq 0.06\)) ceramic samples was prepared by a conventional method. A variation of crystallite size, lattice strain, and dislocation density was observed in Ba\(_2\)Ti\(_3\)O\(_9\) ceramics due to the Sr\(^{2+}\) contents. Their structural properties were investigated by using the X-ray diffraction (XRD) technique. The structural analysis determined the construction of a tetragonal structure with space group \(I4/m\). The average sizes of crystallites decrease from 7.0513 to 1.3979 nm (BST). A scanning electron microscopy (SEM) analysis showed spherical-shaped crystallites. The optical properties were studied by the UV–visible spectra and indicated that the band gap decreases from 2.12 to 1.61 eV with an increase in the Sr\(^{2+}\) concentration. The relative permittivities and quality factors of the samples were calculated at different frequencies. The overall findings in this research study may help in the applications of dielectric resonators.

**MATERIALS AND EXPERIMENTAL METHODS**

Solid solutions of (\(Ba_{1-x}Sr_x\))\(_2\)Ti\(_3\)O\(_9\) (0.00 \(\leq x \leq 0.06\)) ceramics were prepared by a conventional (mixed oxide) route. High-grade raw materials were obtained from Aldrich and Sigma: BaCO\(_3\) (99.9%, Aldrich), TiO\(_2\) (99.9%, Sigma), and SrCO\(_3\) (99.9%, Aldrich) were weighed accordingly and blended as per their stoichiometric ratios. The raw material mixture was dried-ground and mixed by horizontal ball milling for 1 day. After the milling process, the mixture was placed in an alumina crucible and then calcined at 850 °C in a furnace at a heating rate of 5 °C/min for 4 h. The prepared calcined powder was ground manually by using a pestle and mortar for 2.0 h to avoid agglomerations. The soft and fine powder was uniaxially pressed at 90 MPa pressure into a cylindrical pellet with 5.0 mm thickness and 10.0 mm diameter by using a manual pellet press machine (CARVER, USA). Then the green pellet was placed in a high-energy furnace for sintering at a temperature of 1020 °C for 4 h in an open atmosphere at a 5 °C/min cooling/heating rate. The Archimedes principle was used to estimate the approximate bulk densities of the samples. The crystalline phases of the sintered (\(Ba_{1-x}Sr_x\))\(_2\)Ti\(_3\)O\(_9\) (0.00 \(\leq x \leq 0.06\)) ceramics was studied by using an X-ray diffractometer (XRD; JDX-03532, JEOL, Japan) with Cu K\(_\alpha\) (\(\lambda = 0.15406\) nm) radiation operated at 40.0 mA and 40.0 kV in 2θ range from 2θ to 80°. The microstructure of the prepared sample was measured by using impedance spectroscopy (Agilent E4991A, 1 \(\times 10^3\) to 3 \(\times 10^6\) Hz).

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**REFERENCES**

1. Sebastian, M. T. Dielectric materials for wireless communication; Elsevier: 2008; pp 12–14.
2. Lee, Y. C.; Leeb, W. H.; Shieuc, F. S. Microwave dielectric properties and microstructures of Ba\(_2\)Ti\(_9\)O\(_{20}\)-based ceramics with \(ZnO\)-\(BZO\) addition. J. Eur. Ceram. Soc. 2005, 25 (15), 3459–3468.
3. Narang, S. B.; Bahel, S. Low loss dielectric ceramics for microwave applications: a review. J. Ceram. Process. Res. 2010, 11 (3), 316–321.
4. Freer, R.; Azough, F. Microstructural engineering of microwave dielectric ceramics. J. Eur. Ceram. Soc. 2008, 28 (7), 1433–1441.
5. Zhou, D.; Pang, L. X.; Wang, D. W.; Li, C.; Jin, B. B.; Reaney, I. M. High permittivity and low loss microwave dielectrics suitable for 5G resonators and low temperature co-fired ceramic architecture. J. Mater. Chem. C 2017, 5 (38), 10094–10098.
6. Phule, P. P.; Risbud, S. H. Low-temperature synthesis and processing of electronic materials in the BaO-TiO\(_2\) system. J. Mater. Sci. 1990, 25 (2), 1169–1183.
7. Lavat, A. E.; Grasselli, M. C. Synthesis and characterization of ceramic materials based on the system Mg\(_O\)-CaO-TiO\(_2\) from Dolomite. Procedia Mater. Sci. 2015, 8, 162–171.
8. Idles, D. M.; Bell, A. J.; Moulson, A. J. Relationships between dopants, microstructure and the microwave dielectric properties of ZrO\(_2\)-TiO\(_2\)-SnO\(_2\) ceramics. J. Mater. Sci. 1992, 27 (23), 6303–6310.
(9) Choi, Y. J.; Park, J. H.; Ko, W. J.; Park, J. H.; Nahm, S.; Park, J. G. Low temperature sintering of BaTiO3-based middle-k dielectric composition for LTCC applications. J. Electroceram. 2005, 14 (2), 157–162.

(10) Cernea, M. Microwave dielectric properties of BaTiO3-Nd2O3, BaTiO3-Sm2O3 and BaTi4O9-WO3 ceramics. J. Optoelectron. Adv. Mater. 2007, 9 (12), 3790–3794.

(11) Zaman, A.; Uddin, S.; Mehbboob, N.; Tirth, V.; Algahtani, A.; Abbas, M.; Mushqaq, M.; Ali, A.; Sultana, F.; Alhubeiti, K.; et al. Structural elucidation, electronic and microwave dielectric properties of Ca(SnxTi1-x)O3 (0 ≤ x ≤ 0.8). Lead-Free Ceramics. ACS Omega 2022, 7 (5), 4667–4676.

(12) O’Bryan, H. M., Jr.; Thomson, J. Jr. Phase equilibria in the TiO2-rich region of the system BaO-TiO2. J. Am. Ceram. Soc. 1974, 57 (12), 522–526.

(13) Jung, S. W.; Lee, J. H.; Kim, J. Y.; Lee, H. Y.; Cho, S. H. Phase development and microwave dielectric properties of BaO-xSm2O3-xTiO2 (x= 0–1.25) ceramics. Mater. Chem. Phys. 2003, 79 (2–3), 282–285.

(14) Purohit, R. D.; Tyagi, A. K. Synthesis of monophasic Ba2Ti9O20 through gel combustion. J. Mater. Chem. 2002, 12 (4), 1218–1221.

(15) Ji, C.; Wang, J.; Bai, X.; Hao, M.; Chen, G.; He, J.; Fu, C. Effects of MnO2 and WO3 co-doping and sintering temperature on microstructure, microwave dielectric properties of Ba2Ti9O20 micro-waves. Ceram. Int. 2022, 48 (8), 10713–10720.

(16) Shannon, R. D. Revised effective ionic radii and systematic studies of interatomic distances in halides and chalcogenides. Acta Crystallogr., Sect. A: Cryst. Phys., Diffr., Theor. Gen. Crystallogr. 1976, 32 (5), 751–767.

(17) Chu, L. W.; Hsiue, G. H.; Lin, I. N.; Chen, Y. C. Novel reaction mechanism for the synthesis of Ba2Ti9O20 materials prepared from nano-sized oxides. Nanotechnology 2006, 17 (1), 185.

(18) Zaman, A.; Uddin, S.; Mehbboob, N.; Ali, A.; Ahmad, A.; Bashir, K. Effect of Zr+4 on the structural and microwave dielectric properties of CaTiO3 ceramics. Ferroelectrics 2021, 577 (1), 143–152.

(19) Zaman, A.; Uddin, S.; Mehbboob, N.; Ali, A. Structural investigation and improvement of microwave dielectric properties in Ca(HxTi1-x)O3 ceramics. Phys. Scr. 2021, 96 (2), 025701.

(20) Lin, W. Y.; Speyer, R. F. Dielectric properties of microstructure-controlled Ba2Ti9O20 resonators. J. Am. Ceram. Soc. 1999, 82 (2), 325–330.

(21) Zaman, A.; Uddin, S.; Mehbboob, N. Synthesis and Microwave Dielectric Characterization of Ca1-xSmTiO3, Low-Loss Ceramics. Iran. J. Sci. Technol., Trans. A: Sci. 2021, 45 (1), 367–371.

(22) O’Bryan, H. M., Jr.; Thomson, J. Jr. Phase equilibria in the TiO2-rich region of the system BaO-TiO2. J. Am. Ceram. Soc. 1974, 57 (12), 522–526.

(23) Lei, S.; Fan, H.; Ren, X.; Fang, J.; Ma, L.; Tian, H. Microstructure, phase evolution and interfacial effects in a new Zn0.9Mg0.1TiO3-ZnNb2O6 ceramic system with greatly induced improvement in microwave dielectric properties. Scr. Mater. 2018, 146, 154–159.

(24) Luo, T.; Yang, Q.; Yu, H.; Liu, J. Formation mechanism and microstructure evolution of Ba2Ti9O20 ceramics by reaction sintering method. J. Am. Ceram. Soc. 2020, 103 (2), 1079–1087.

(25) Lin, W. Y.; Speyer, R. F.; Hackenberger, W. S.; Shront, T. R. Microwave properties of Ba2Ti9O20 doped with zirconium and tin oxides. J. Am. Ceram. Soc. 1999, 82 (5), 1207–1211.

(26) Yang, X.; Li, B.; Zhang, C.; Wang, S.; Liu, K.; Zou, C. Fabrication and properties of porous silicon nitride wave-transparent ceramics via gel-casting and pressureless sintering. Mater. Sci. Eng.: A 2016, 663, 174–180.

(27) Shelhar, M. B.; Jadhav, P. A.; Chougule, S. S.; Mallapur, M. M.; Chougule, B. K. Structural and electrical properties of nickel cadmium ferrites prepared through self-propagating auto combustion method. J. Alloys Compd. 2009, 476 (1–2), 760–764.

(28) Murugesan, C.; Ugendar, K.; Okrasa, L.; Shen, J.; Chandrasekaran, G. Zinc substitution effect on the structural, spectroscopic and electrical properties of nanocrystalline MnFe2O4 spinel ferrite. Ceram. Int. 2021, 47 (2), 1672–1685.