

# Effect of Microwave Sintering on the Microstructure and Dielectric Properties of $\text{Bi}_2\text{O}_3$ -Doped $(\text{Ba}_{0.6}\text{Sr}_{0.4})(\text{Ti}_{0.94}\text{Cu}_{0.06})\text{O}_3$ Ceramics

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## ABSTRACT

The effects of microwave sintering on the sintering behaviour, microstructure and dielectric properties of  $\text{Bi}_2\text{O}_3$ -doped  $(\text{Ba}_{0.6}\text{Sr}_{0.4})(\text{Ti}_{0.94}\text{Cu}_{0.06})\text{O}_3$  (BSTC) ceramics were investigated. The microstructure and dielectric properties of a BSTC ceramic were also studied given different amounts of  $\text{Bi}_2\text{O}_3$  doping. Microwave heating with sintering temperatures below  $1000^\circ\text{C}$  significantly improves the densification of  $\text{Bi}_2\text{O}_3$ -doped BSTC ceramics. The BSTC ceramic with 1 wt%  $\text{Bi}_2\text{O}_3$  addition sintered at  $950^\circ\text{C}$  in air for 30 min exhibited dielectric properties of  $\epsilon_r = 3756$ , dielectric loss of  $\tan\delta = 7 \times 10^{-3}$  and bulk density > 96% of theoretical density.

## KEYWORDS

$(\text{Ba}_{0.6}\text{Sr}_{0.4})(\text{Ti}_{0.94}\text{Cu}_{0.06})\text{O}_3$  (BSTC); Dielectric Properties; Microwave Sintering; Microstructures

## 1. Introduction

It is well known that barium and strontium titanates ( $\text{BaTiO}_3$  and  $\text{SrTiO}_3$ ) can be formed by the solid solutions because of their identical crystal structures and the comparable ionic radii of  $\text{Ba}^{2+}$  and  $\text{Sr}^{2+}$  [1-4]. This material is very promising for practical applications, such as phase shifter, delay lines, tunable filters, steerable antennas, etc. [5-7]. However, an ideal BST ( $\text{BaSrTiO}_3$ ) system should exhibit the following characteristics: high dielectric constant ( $\epsilon_r$ ), low dissipation factor ( $\tan\delta$ ), large tunability and a low temperature dependence [5,6,8].

Pure BST ceramics have to be sintered at  $\sim 1350^\circ\text{C}$ , a temperature range much higher than the melting point of conductors such as Ag ( $961^\circ\text{C}$ ) or Cu ( $1083^\circ\text{C}$ ). Therefore, only platinum or refractory metals can be used as inner conductors [7]. These materials are expensive, their electrical performance is poor, and the cost-effective fabrication of high-quality components is difficult. Several methods are commonly used for lowering the sintering

temperature of dielectric ceramics, such as: the addition of glass; chemical pre-treatment and processing of ceramic powders; and the addition of small particles of starting materials [9-11]. Of these three methods, liquid-phase sintering with glass addition is generally known to be the most effective and the least expensive way of achieving high density sintered ceramics [12,13].

Researchers have already reported attempts to decrease the sintering temperature of BST to  $900^\circ\text{C}$  so that it can be used in LTCC (Low temperature cofiring ceramics) applications. Rhim *et al.* [9] reported on the influence of  $\text{B}_2\text{O}_3$  additions on the sintering temperature of commercial  $\text{Ba}_{0.7}\text{Sr}_{0.3}\text{TiO}_3$ ; they showed that the addition of 0.5 wt%  $\text{B}_2\text{O}_3$  reduces the sintering temperature to  $1150^\circ\text{C}$ , while the dielectric ferroelectric properties remain unchanged. Valant *et al.* [14] reported on the influence of  $\text{Li}_2\text{O}$  additions on the sintering temperature of  $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ ; they showed that an addition of only 0.4 wt%  $\text{Li}_2\text{O}$  to  $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$  powder was able to reduce the sintering temperature to  $\leq 900^\circ\text{C}$  and produce ceramics with a relative density of 97%.

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Microwave sintering is a method of internal self-heating through the absorption of microwave power. Therefore, internal microwave sintering can be compared with external sintering by thermal conduction or radiation. Microwave sintering is expected to produce a fine grain, a high degree of uniformity and high densification in ceramics, along with improving electrical and mechanical properties [15].

This experiment is based on (Ba<sub>0.6</sub>Sr<sub>0.4</sub>)(Ti<sub>0.94</sub>Cu<sub>0.06</sub>)O<sub>3</sub> (BSTC) because of its low  $T_c$ , high dielectric constant, relatively low loss tangent, and good tunability. In a previous study [16], the dielectric constant of (Ba<sub>0.6</sub>Sr<sub>0.4</sub>)(Ti<sub>0.94</sub>Cu<sub>0.06</sub>)O<sub>3</sub> ceramics sintered at 1150°C was determined as 4094. However, the sintering temperature of 1150°C is still too high for LTCC applications. To reduce sintering temperature, the Bi<sub>2</sub>O<sub>3</sub> was added in BSTC ceramics as a sintering aid. On the other hand, to decrease the volatility of the bismuth elements during sintering, the microwave process was employed to sinter Bi<sub>2</sub>O<sub>3</sub>-doped BSTC ceramics. In this paper, we examined the effects of microwave sintering on the sintering behaviour, microstructure and dielectric properties of Bi<sub>2</sub>O<sub>3</sub>-doped BSTC ceramics.

## 2. Experimental Procedure

Conventional ceramic fabrication processes were used to prepare the present BSTC samples from commercial powders of BaCO<sub>3</sub>, SrCO<sub>3</sub>, CuO and TiO<sub>2</sub>. The BaCO<sub>3</sub>, SrCO<sub>3</sub>, TiO<sub>2</sub>, and CuO powders were mixed with deionized water for 24 h in a  $\phi$ 2 mm zirconia ball-mill. The mixture was dried, calcined at 1100°C for 6 h in air, and then crushed into a powder. An appropriate amount of Bi<sub>2</sub>O<sub>3</sub> was mixed with the BSTC powders using the same procedure. The powders were mixed with the binder (Polyvinyl alcohol; PVA) additive and then were pressed into disk-shaped specimens. The samples were sintered in air by microwave processing, ramping at 30°C/min. The temperature of the sample was monitored with a type-R thermocouple shielded with platinum foil and grounded to the inner metallic wall of the microwave furnace. The samples were sintered at various temperatures from 900°C to 1000°C, held for 30 min at the peak temperature. Samples were microwave sintered using a single-mode microwave furnace with a cavity of 37 cm  $\times$  34.5 cm  $\times$  33.5 cm. The microwave sintering experiments were conducted in a 2.4 kW, 2.45 GHz. The samples were encased in a microwave susceptor (SiC) located in a thermal insulation package in the microwave chamber. Sintering temperature is measured using an optical pyrometer (President Honor industries Co., Ltd., Taiwan) focused directly onto the samples. The model of optical pyrometer is SH60 which can be applied at 600°C - 1200°C. The pyrometer is directly connected to the

controller and does not influence or interfere with the microwave field distribution within the cavity. However, the traditional metal thermocouples can interfere with the microwave field within the cavity preventing accurate measurements from being made. The optical pyrometer was calibrated at several temperature points using a type B PtRh thermocouple placed in contact with the samples.

The crystalline phases of the sintered ceramics were identified by X-ray diffraction pattern analysis (XRD, Bruker D8A, Germany) using Cu-K $\alpha$  radiation for  $2\theta$  from 20° to 80°. The diffraction spectra were collected at a scan rate of 2.5°/min. Microstructural observation of the sintered ceramics was performed using a scanning electron microscope (SEM, JEOL. JEL-6400 Japan) equipped with energy-dispersive spectroscopy (EDS). The bulk density of the sintered pellets was measured using the Archimedes method. Particle size was measured using a particle size analyzer (Malvern, Mastersizer 2000, UK). The capacitance and dissipation factor were measured at 1MHz and 23°C (HP4278A). The dielectric properties of the samples were measured as a function of temperature using a HP 4284A LCR meter and programmable temperature chamber interfaced to a PC for automated measurements, and samples were measured at temperatures ranging from -55°C to 125°C.

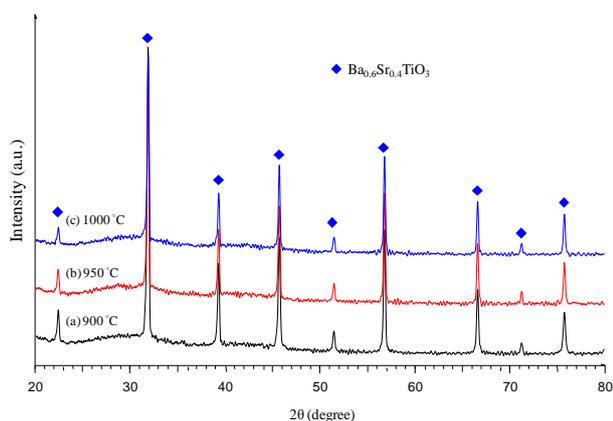
## 3. Results and Discussion

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### 3.1. Phase Evolution in the Sintered Ceramics

This study aims to find a way to reduce the sintering temperature of BSTC. The influence of Bi<sub>2</sub>O<sub>3</sub> on the microstructural characteristics and dielectric properties was studied. The interaction of Bi<sub>2</sub>O<sub>3</sub> with BSTC after microwave heating was investigated using X-ray diffraction. **Figure 1** shows the X-ray diffraction spectra for the as-sintered BSTC ceramics with 1 wt% Bi<sub>2</sub>O<sub>3</sub> addition, sintered in air for 30 min at temperatures ranging from 900 to 1000°C. All the samples are composed of a single phase, indicating that Bi<sub>2</sub>O<sub>3</sub> did not form a crystalline phase for composition doped with 1.0 wt% Bi<sub>2</sub>O<sub>3</sub>. It has been reported that the BST phase has high dielectric con-



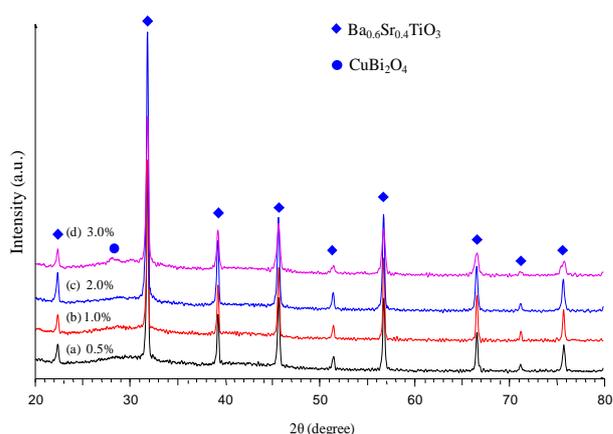
**Figure 1.** X-ray diffraction spectra of BSTC ceramics doped with 1 wt%  $\text{Bi}_2\text{O}_3$  sintered at (a) 900 °C, (b) 950 °C and (c) 1000 °C.

stants, low dielectric losses, high tunability [5,8,17] and could be suitable for use in dielectric resonators at microwave frequencies. **Figure 2** shows the XRD patterns of the BSTC ceramics with different  $\text{Bi}_2\text{O}_3$  addition at 950 °C. For 0.5 wt%, 1 wt% and 2 wt%  $\text{Bi}_2\text{O}_3$  doped BSTC ceramics, as shown in **Figures 2(a)-(c)**, it was determined that there is a single crystalline phase of BST present in the specimen. For BSTC ceramics with 3 wt%  $\text{Bi}_2\text{O}_3$  addition, as shown in **Figure 2(d)**, it was found that the major crystalline phase is the same as BSTC ceramics doped with lower  $\text{Bi}_2\text{O}_3$  content ( $\leq 2$  wt%). However,  $\text{CuBi}_2\text{O}_4$  of minor impurity phases was detected in the specimens.

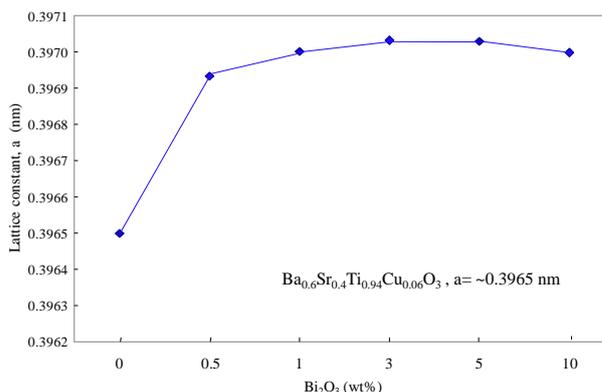
**Figure 3** shows the variation of the lattice parameter as a function of the addition of  $\text{Bi}_2\text{O}_3$  sintered at 950 °C. With increasing levels of  $\text{Bi}_2\text{O}_3$  addition, the lattice constant initially increased suddenly, then kept approximately a constant. This result shows that a small amount of  $\text{Bi}_2\text{O}_3$  addition led to  $\text{Cu}^{2+}$  precipitation from the grain in BSTC materials. When Cu substitutes into Ti sites in the perovskite  $\text{ABO}_3$  structure, it creates lattice strain and oxygen vacancies [18]. Therefore, the lattice parameter,  $a$ , of BSTC is smaller than the lattice parameter of BST.

### 3.2. Microstructural Evolution in the Sintered Ceramics

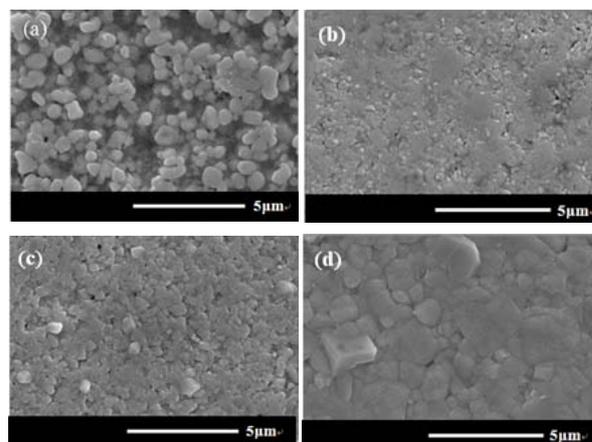
SEM micrographs of the BSTC specimens sintered at 950 °C with 0, 0.5, 1, and 3 wt% of  $\text{Bi}_2\text{O}_3$  are shown in **Figures 4(a)-(d)**, respectively. The images show that significant densification of BSTC ceramics with  $\text{Bi}_2\text{O}_3$  dopant occurred. It is widely accepted that pure BSTC ceramic has to be sintered at 1150 °C for several hours [16]. Therefore these results show that a small amount of  $\text{Bi}_2\text{O}_3$  dopant can increase the density of BSTC ceramics with microwave firing at 950 °C. These SEM images confirm that the ceramics are sintered to a high density. The



**Figure 2.** X-ray diffraction spectra for BSTC ceramics sintered at 950 °C and doped with various  $\text{Bi}_2\text{O}_3$  concentrations: (a) 0.5 wt%; (b) 1.0 wt%; (c) 2.0 wt% and (d) 3.0 wt%.



**Figure 3.** The variation of lattice parameter with amounts of  $\text{Bi}_2\text{O}_3$  added into BSTC ceramics.



**Figure 4.** SEM micrographs of the BSTC ceramics with (a) 0 wt%, (b) 0.5 wt%, (c) 1.0 wt% and (d) 3.0 wt%  $\text{Bi}_2\text{O}_3$  addition and sintered at 950 °C for 30 min.

relative density of BSTC ceramics is discussed below. The effect of microwave sintering temperature on microstructure was also observed. SEM micrographs of the

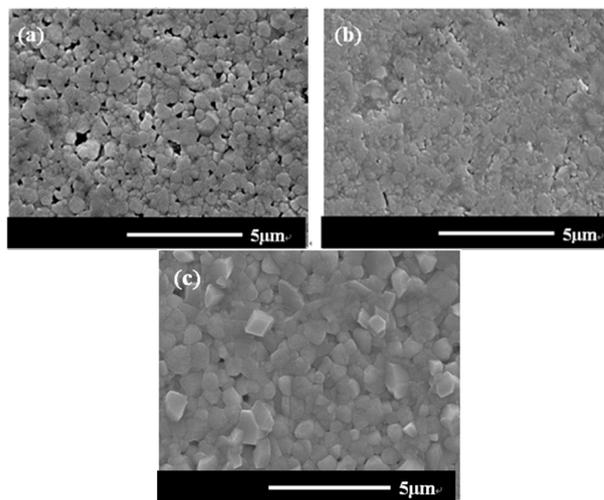
polished surfaces of the 1 wt%  $\text{Bi}_2\text{O}_3$ -doped BSTC specimens are shown in **Figures 5(a)-(c)**, for sintering at 900, 950, and 1000°C, respectively. In **Figure 5(a)**, it can be seen that the BSTC ceramic with 1 wt%  $\text{Bi}_2\text{O}_3$  addition sintered with microwave heating at 900°C is rather porous, therefore the densification of the ceramic is insufficient. As the sintering temperature increases, the size and number of voids in the sintered specimen is considerably reduced, as shown in **Figures 5(b)** and **(c)**.

For BSTC with 3 wt%  $\text{Bi}_2\text{O}_3$  addition, we observed a microstructure with a significant amount of abnormal grains as shown in **Figures 4(d)** and **5(c)**. According to the XRD spectra shown in **Figure 2**, the composition of the secondary phase should be  $\text{CuBi}_2\text{O}_4$  at  $\text{Bi}_2\text{O}_3 > 2$  wt%. Energy dispersive spectroscopy (EDS) was used to clarify the issue, and the results of BSTC ceramic with 3 wt%  $\text{Bi}_2\text{O}_3$  addition sintered at 950°C are shown in **Figure 6**. Compared with the matrix phase (**Figure 6(a)**), the abnormal grains have higher signal intensities in Cu and Bi (**Figure 6(b)**). Moreover, the X-ray diffraction analysis in the previous section clearly shows that the formation of the  $\text{CuBi}_2\text{O}_4$  phase was preferred at  $\text{Bi}_2\text{O}_3 > 2$  wt%. It is therefore believed that the abnormal grains are composed of  $\text{CuBi}_2\text{O}_4$ .

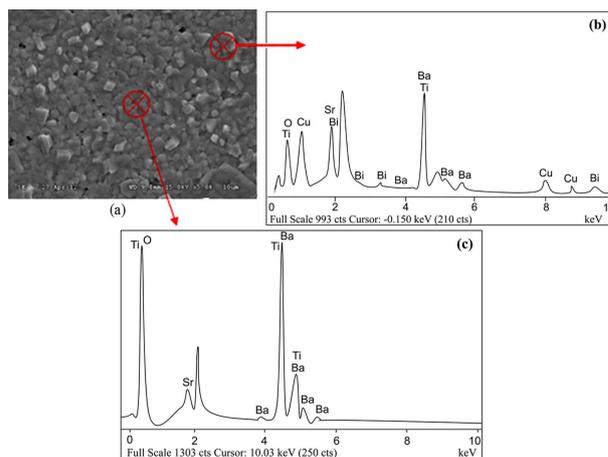
### 3.3. Density and Dielectric Properties of the Sintered Ceramics

As described in the experimental section, the BSTC ceramics with different  $\text{Bi}_2\text{O}_3$  additions were sintered in air at temperatures ranging from 900°C to 1000°C for 30 min. **Figure 7** shows the bulk density of the BSTC ceramics as functions of  $\text{Bi}_2\text{O}_3$  content and sintering temperature. The bulk density of the BSTC increased at higher sintering temperatures and  $\text{Bi}_2\text{O}_3$  addition. The theoretical density of the  $\text{B}_{0.6}\text{Sr}_{0.4}(\text{Ti}_{0.94}\text{Cu}_{0.06})\text{O}_3$  ceramics is approximately  $5.69 \text{ g/cm}^3$  (based on a  $\text{CuO}$  density of  $6.3 \text{ g/cm}^3$ ) [14]. The BSTC ceramics with 0.5 wt%  $\text{Bi}_2\text{O}_3$  addition can be sintered to over 96% of the theoretical density (*i.e.*  $5.46 \text{ g/cm}^3$ , at 950°C for 30 min). However, the influence of sintering temperature on the bulk density of the BSTC ceramics depends on the amounts of  $\text{Bi}_2\text{O}_3$  addition, so the microwave sintering temperature of BSTC can be reduced to 950°C by increasing the glass forming content and adopting correct BSTC ceramic composition.

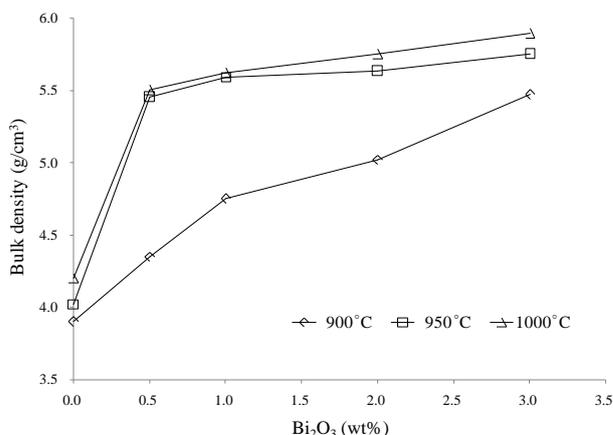
It is well known, pure BSTC ceramic has to be sintered at  $\sim 1150^\circ\text{C}$  for several hours, therefore BSTC ceramics with a small amount of  $\text{Bi}_2\text{O}_3$  addition using microwave heating can increase the density of BSTC ceramics at 950°C. Related results reported by other investigators [13,19] show that the  $\text{Bi}_2\text{O}_3$  assists in the densification of the BST dielectrics through liquid-phase sintering. It is also interesting to note that the sintered ce-



**Figure 5.** SEM micrographs of the BSTC ceramics with 1 wt%  $\text{Bi}_2\text{O}_3$  addition sintered at (a) 900°C (b) 950°C and (c) 1000°C for 30 min.



**Figure 6.** The BSTC ceramic with 3 wt%  $\text{Bi}_2\text{O}_3$  addition sintered at 950°C, where: (a) morphology; (b) EDS of secondary phase; (c) EDS of the matrix.

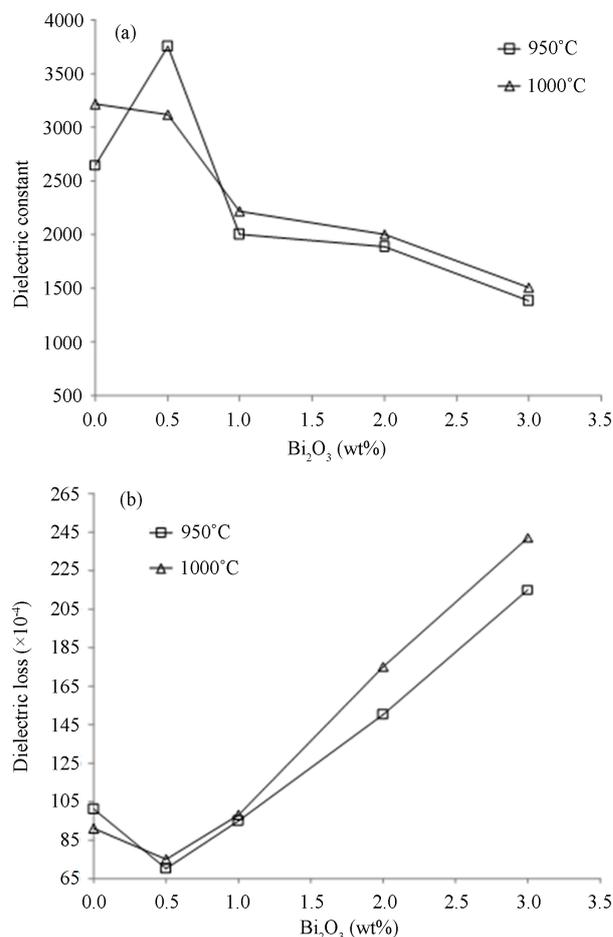


**Figure 7.** Bulk densities of BSTC ceramics with different  $\text{Bi}_2\text{O}_3$  addition as a function of sintering temperature.

amics with the highest bulk density is the ceramics with the greatest amount of Bi<sub>2</sub>O<sub>3</sub> addition at 1000°C. In fact, the addition of 3 wt% Bi<sub>2</sub>O<sub>3</sub> to the BSTC ceramics that were sintered at 1000°C has resulted in the highest density among the sintered specimens. The reason for this is that overdoped Bi<sub>2</sub>O<sub>3</sub> did not become volatile or form an appropriate amount of the liquid phase; it remained in the specimens and formed a secondary phase. According to SEM morphology and XRD analysis, the secondary phase of CuBi<sub>2</sub>O<sub>4</sub> increased significantly with greater Bi<sub>2</sub>O<sub>3</sub> additions. Hasegawa *et al.* [20] reported that the CuBi<sub>2</sub>O<sub>4</sub> with melting point of 600°C and density of 8.5 g/cm<sup>3</sup> was effective for low-temperature sintering of Ba-TiO<sub>3</sub>. Therefore, the bulk density of the BSTC ceramic can be increased when more CuBi<sub>2</sub>O<sub>4</sub> forms in the samples.

The dielectric constant ( $\epsilon_r$ ) of Bi<sub>2</sub>O<sub>3</sub>-doped BSTC ceramics as functions of microwave sintering temperatures and amount of Bi<sub>2</sub>O<sub>3</sub> addition was measured at 1 MHz at ambient temperature with the results shown in **Figure 8(a)**. The dielectric constant of the BSTC ceramics increased with sintering temperature. The dielectric constants of the samples increased with the amount of Bi<sub>2</sub>O<sub>3</sub> addition, and reached a maximum value at 0.5 wt% at 950°C sintering, then gradually decreased with increasing amounts of Bi<sub>2</sub>O<sub>3</sub>. In addition, the dielectric constant of the sample with 0.5 wt% Bi<sub>2</sub>O<sub>3</sub> sintered at 950°C was ~3756, higher than that of the sample with 1 wt% Bi<sub>2</sub>O<sub>3</sub> (~2002). The reason for this is due to the decrease of lattice strain as shown in **Figure 3**, a small amount of Bi<sub>2</sub>O<sub>3</sub> addition led to Cu<sup>2+</sup> precipitation from the grain in BSTC materials. When Cu substitutes into Ti sites in the perovskite ABO<sub>3</sub> structure, it creates lattice strain and oxygen vacancies. Preethi *et al.* [21] reported that the distortion of the crystalline lattice with respect to a decrease in the ionic radii might be also the reason for the increase in dielectric constant. **Figure 8(b)** shows the dielectric loss ( $\tan\delta$ ) of the BSTC ceramics at 1 MHz as functions of sintering temperatures and amount of Bi<sub>2</sub>O<sub>3</sub> addition. Similarly, the evolution of the dielectric loss of Bi<sub>2</sub>O<sub>3</sub>-doped BSTC ceramics reached a minimum at 0.5 wt% and 950°C sintering temperature, and then increased gradually with increasing Bi<sub>2</sub>O<sub>3</sub> content. However, the secondary phases exist for BSTC ceramics doped with Bi<sub>2</sub>O<sub>3</sub>, a decrease in dielectric constant is mainly associated with the formation of the second phase, CuBi<sub>2</sub>O<sub>4</sub> phase, that has a lower dielectric constant in comparison with that of BSTC.

Measurement of the electrical properties over a temperature range of -55°C - 125°C enabled the Curie peak at the Curie temperature, T<sub>c</sub>, to be observed and also gave an idea of the possible effects of temperature variation (*i.e.* in the region of room temperature which is im-

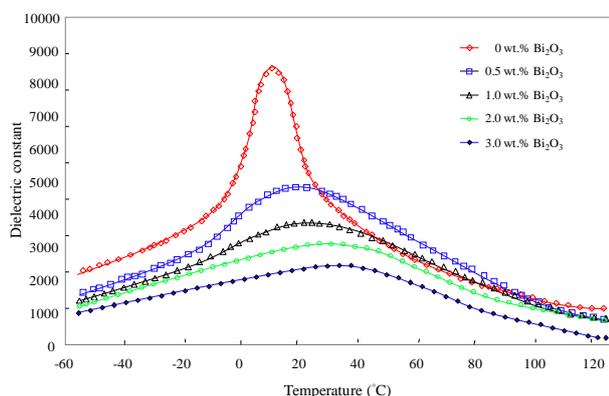


**Figure 8.** The dielectric constant and dielectric loss of BSTC ceramics with different Bi<sub>2</sub>O<sub>3</sub> addition as a function of sintering temperature.

portant for most applications) [22]. The dielectric constant as a function of temperature at 1 MHz for BSTC with different levels Bi<sub>2</sub>O<sub>3</sub> doping are shown in **Figure 9**, all the samples were sintered at 950°C. The dielectric constant decreased with increasing amounts of Bi<sub>2</sub>O<sub>3</sub>. The reason for this is that larger amounts of Bi<sub>2</sub>O<sub>3</sub> led to the formation of a second phase. So it will be helpful to decrease the distortion of the crystal lattice and release the inner stress caused by precipitating with CuBi<sub>2</sub>O<sub>4</sub> secondary phases, result in the decrease of dielectric constant.

#### 4. Conclusion

The effect of the addition of Bi<sub>2</sub>O<sub>3</sub> and microwave sintering on phase evolution, microstructure and dielectric properties of B<sub>0.6</sub>Sr<sub>0.4</sub>(Ti<sub>0.94</sub>Cu<sub>0.06</sub>)O<sub>3</sub> ceramics has been investigated. It was discovered that adding Bi<sub>2</sub>O<sub>3</sub> to BSTC ceramics by microwave heating can lower the sintering temperature from 1150°C to 950°C and in-



**Figure 9.** Temperature dependence of permittivity and dielectric loss of BSTC ceramics with different  $\text{Bi}_2\text{O}_3$  addition. Permittivity and dielectric loss were measured at 1 MHz.

creases the bulk density of the sintered ceramics. An X-ray diffraction examination of the products indicated that they consist mainly of a  $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$  crystalline phase with a  $\text{CuBi}_2\text{O}_4$  as secondary phase, which formed as a result of  $\text{Bi}_2\text{O}_3$  addition. A BSTC ceramics formed by adding 0.5 wt%  $\text{Bi}_2\text{O}_3$  and microwave heating at  $950^\circ\text{C}$  in air for 2 h gives a permittivity,  $\epsilon_r = 3756$ ,  $\tan\delta = 7 \times 10^{-3}$  and a bulk density >96% of theoretical.

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