



Substituting La^{3+} with Sr^{2+} to improve microwave dielectric properties of $\text{La}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics

Yih-Chien Chen*, Wei-Yu Hsu

Department of Electrical Engineering, Lunghwa University of Science and Technology, Gueishan Shiang, Taoyuan County, Taiwan

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ABSTRACT

The microwave dielectric properties of $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics were examined with a view to their exploitation for mobile communication. The $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics were prepared by the conventional solid-state method with various sintering temperatures. The X-ray diffraction peaks of the $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics shifted to lower angles as x increased from 0 to 0.05. The X-ray diffraction patterns of the $\text{La}_{2.98/3}\text{Sr}_{0.01}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics revealed no significant variation of phase with sintering temperatures. A maximum density of 6.56 g/cm^3 was obtained for $\text{La}_{2.98/3}\text{Sr}_{0.01}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramic, sintered at 1550°C for 4 h. A dielectric constant (ϵ_r) of 20.0 and a product of quality factor and resonant frequency ($Q \times f$) of 57,100 GHz and a temperature coefficient of resonant frequency (τ_f) of $-77\text{ ppm}/^\circ\text{C}$ were obtained for $\text{La}_{2.98/3}\text{Sr}_{0.01}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics that were sintered at 1550°C for 4 h.

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1. Introduction

Many modern communication systems, such as mobile radio and wireless communications, use resonators, filters, and antennas. However, these microwave devices have limitations in size, gain, efficiency, and temperature stability, which are imposed by the microwave dielectric material. Microwave materials that are to be used in microwave devices must have three dielectric properties – a high dielectric constant, a high quality factor, and a near-zero temperature coefficient of resonant frequency, which enable small size, low loss and high temperature stability, respectively [1,2].

The advantages of using complex perovskite ceramics $A(\text{B}'_{0.5}\text{B}''_{0.5})\text{O}_3$ ($A = \text{Me}^{2+}, \text{Me}^{3+}$; $\text{B}' = \text{Me}^{2+}, \text{Me}^{3+}$; $\text{B}'' = \text{Me}^{4+}, \text{Me}^{5+}, \text{Me}^{6+}$) are reportedly associated with their excellent dielectric properties at microwave frequencies [3–5]. $\text{Ln}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ ceramics combine a high dielectric constant ($\epsilon_r \sim 22$ –27), a high quality factor ($Q \sim 3770$ –7550 at 10 GHz), and an adjustable temperature coefficient of resonant frequency.

The dielectric constant and temperature coefficient of resonant frequency of $\text{La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ ceramics can be improved by introducing another compound with positive temperature coefficient of resonant frequency to form a solid solution. $0.55\text{La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ – 0.45SrTiO_3 ceramics with 0.25 wt% B_2O_3 sin-

tered at 1475°C for 4 h in air exhibited optimum microwave dielectric properties of $\epsilon_r = 46.3$ and $Q \times f = 34,000\text{ GHz}$, and $\tau_f = -0.12\text{ ppm}/^\circ\text{C}$ [6]. $0.6\text{La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ – $0.4(\text{La}_{0.5}\text{Na}_{0.5})\text{TiO}_3$ ceramics with 1 wt% B_2O_3 addition possesses a ϵ_r of 36.0, a $Q \times f$ value of 25,500 GHz, and a τ_f of $-5\text{ ppm}/^\circ\text{C}$ sintering at 1475°C [7]. $0.6\text{La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ – $0.4\text{Ca}_{0.6}\text{La}_{0.8/3}\text{TiO}_3$ ceramics with 0.25 wt% ZnO addition possesses a ϵ_r of 43.6, a $Q \times f$ value of 48,000 GHz, and a τ_f of $-1\text{ ppm}/^\circ\text{C}$ sintering at 1475°C [8]. $0.6\text{La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ – $0.4\text{Ca}_{0.6}\text{La}_{0.8/3}\text{TiO}_3$ ceramics with 1 wt% CuO addition possesses a ϵ_r of 45.5, a $Q \times f$ value of 44,600 GHz, and a τ_f of $2\text{ ppm}/^\circ\text{C}$ sintering at 1450°C [9]. Since the ionic radius of Sn^{4+} (0.074 nm) is similar to that of Ti^{4+} (0.064 nm), the Ti^{4+} ion can be substituted by the Sn^{4+} ion to form $\text{Ln}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics [10]. The dielectric constant of $\text{La}(\text{Mg}_{0.5}\text{Ti}_{0.5-x}\text{Sn}_x)\text{O}_3$ perovskite ceramics was found to gradually decrease from 28.4 to 19.7 with an increase in tin concentration, whereas the temperature coefficient of resonant frequency decreased from -68 to $-84\text{ ppm}/^\circ\text{C}$ [11]. A dielectric constant of 15.6 and a $Q \times f$ of 30,600 GHz were obtained for $\text{La}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics that were sintered at 1500°C for 4 h [12]. A liquid phase flux such as B_2O_3 and CuO was added to lower the sintering temperature of $\text{La}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics. A dielectric constant of 19.7 and a $Q \times f$ of 43,300 GHz were obtained for $\text{La}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics with 0.5 wt% CuO additive sintered at 1550°C for 4 h [13]. Additionally, a dielectric constant of 19.7, and a $Q \times f$ of 45,000 GHz were obtained for $\text{La}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics with 0.5 wt% B_2O_3 additive sintered at 1500°C for 4 h [14]. $\text{BaLa}_4\text{Ti}_4\text{O}_{15}$ compounds were found to have good microwave dielectric properties com-

* Corresponding author. Tel.: +886 2 8209 3211; fax: +886 2 8209 9728.

E-mail addresses: EE049@mail.lhu.edu.tw, ycchenncku@yahoo.com.tw (Y.-C. Chen).

pared with that of $\text{CaLa}_4\text{Ti}_4\text{O}_{15}$ compounds [15]. The fact that the ionic radius of Ba^{2+} ions (0.135 nm) is larger than that of La^{3+} ions (0.103 nm), motivates this study of the effect of the substitution of La^{3+} by Sr^{2+} to form $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics. In this paper, $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ were synthesized and some of the La^{3+} ions were substituted with Sr^{2+} ions to improve their microwave dielectric properties. Since the ionic radius of Sr^{2+} ions (0.118 nm) is larger than that of La^{3+} ions (0.103 nm). Moreover, the effect of the sintering temperature on the microwave dielectric properties of $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics was studied. $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics were synthesized herein by the conventional mixed-oxide method and demonstrated proven to have better microwave dielectric properties than $\text{La}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics. The dielectric properties of the $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics at microwave frequencies were found to vary with the degree of Sr^{2+} substitution and sintering temperatures. These various microwave dielectric properties were analyzed by densification, X-ray diffraction (XRD) patterns, and observation of their microstructures.

2. Experimental procedures

The starting raw chemicals were high-purity La_2O_3 (99.99%), SrCO_3 (99.9%), MgO (98.0%), and SnO_2 (99.0%) powders. The prepared composition was $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$. Specimens were prepared using the conventional mixed-oxide method. The starting materials were stoichiometrically weighed after drying La_2O_3 at 1000°C for 4 h to remove moisture content. The raw material was ball-milled in alcohol for 12 h, dried, and then calcined at 1200°C for 4 h. The calcined powder was re-milled for 12 h using PVA solution as a binder. The obtained fine powder was then crushed into a finer powder through a sieve with a 200 mesh. The obtained fine powder was then axially pressed at 2000 kg/cm^2 into pellets with a diameter of 11 mm and a thickness of 6 mm. The specimens thus obtained were then sintered at temperatures of $1450\text{--}1600^\circ\text{C}$ for 4 h in air. Both the heating rate and the cooling rate were set to 10°C/min .

After sintering, the phases of the samples were investigated by X-ray diffraction. An X-ray Rigaku D/MAX-2200 was used with $\text{CuK}\alpha$ radiation (at 30 kV and 20 mA) and a graphite monochromator in the 2θ range of $20\text{--}80^\circ$. Scanning electron microscopy (SEM; JEOL JSM-6500F) and energy dispersive X-ray spectrometer (EDS) were utilized to examine the microstructures of the specimens. The apparent densities of the specimens were measured by the liquid Archimedes method using distilled water as the liquid. The microwave dielectric properties of the specimens were measured by the postresonator method that was developed by Hakki and Coleman [16]. The postresonator method employed a specimen in the form of a cylinder of diameter D and length L . The specimens used for making microwave dielectric property measurements had an aspect ratio D/L of about 1.6, which is in the permitted range determined by Kobayashi and Katoh [17]. The cylindrical resonator was sandwiched between two conducting plates. Two small antennas were positioned in the vicinity of the specimen to couple the microwave signal power into or out of the resonator. The other ends of the antennas were connected to an Agilent E5071C network analyzer. The resonance characteristics depended on the size and microwave dielectric properties of the specimen. The microwave energy was coupled using electric-field probes. The TE_{011} resonant mode was optimal for obtaining the dielectric constant and the loss factor of the specimen. The Agilent E5071C network analyzer was used to identify the TE_{011} resonant frequency of the dielectric resonator, and the dielectric constant and quality factor were calculated. The technique for measuring τ_f was the same as that for measuring the dielectric constant. The test cavity was placed in a chamber and the temperature was increased from 25 to 75°C . The τ_f value (ppm/ $^\circ\text{C}$) can be determined by noting the change in resonant frequency,

$$\tau_f = \frac{f_2 - f_1}{f_1(T_2 - T_1)}, \quad (1)$$

where f_1 and f_2 represent the resonant frequencies at T_1 and T_2 , respectively.

3. Results and discussion

Fig. 1 shows the X-ray diffraction patterns of $\text{La}_{2.98/3}\text{Sr}_{0.01}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics that were sintered at $1450\text{--}1600^\circ\text{C}$ for 4 h. Clearly, $\text{La}_{2.98/3}\text{Sr}_{0.01}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ is the main crystalline phase, which is accompanied by small amounts of $\text{La}_2\text{Sn}_2\text{O}_7$ as the second phases. All of the peaks were indexed based on the cubic perovskite unit cell. A series of extra peaks were observed to correspond to superlattice reflections. All of the superlattice reflections were indexed using half-integer

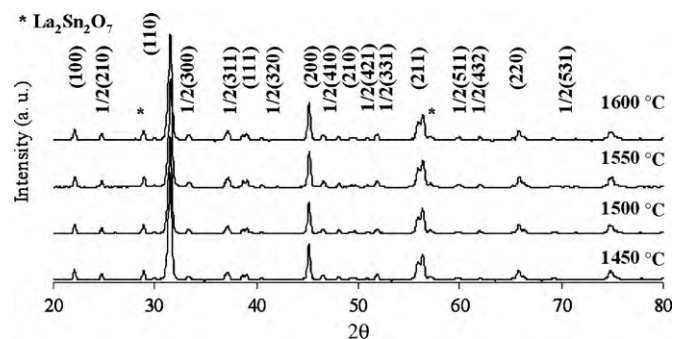


Fig. 1. X-ray diffraction patterns of $\text{La}_{2.98/3}\text{Sr}_{0.01}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ specimens sintered at $1450\text{--}1600^\circ\text{C}$ for 4 h.

Miller indices. According to Glazer, the superlattice reflections, with specific combinations of odd (o) and even (e) Miller indices, point to definite deviations of the structure from the undistorted cubic structure, such as octahedral in-phase tilting (ooe, oeo, eoo), anti-phase tilting (ooo, $h+k+l>3$), chemical ordering (ooo) and anti-parallel displacement of A-cations (eoo, eoe, oee) [18]. The extra $(1/2(210))$, $(1/2(300))$, $(1/2(320))$, $(1/2(410))$, $(1/2(421))$ and $(1/2(432))$ peaks indicate that A-site cation displacement and the $(1/2(311))$, $(1/2(331))$, $(1/2(511))$ and $(1/2(531))$ peaks indicate that anti-phase tilting. The X-ray diffraction patterns of the $\text{La}_{2.98/3}\text{Sr}_{0.01}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics do not significantly vary with sintering temperature. Fig. 2 shows the X-ray diffraction patterns of $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics with x varied from 0 to 0.05 in the 2θ range of $28\text{--}38^\circ$, following sintering at 1550°C for 4 h. The diffraction peaks of $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics shifted to lower angles as x increased, perhaps because the ionic radius of Sr^{2+} ions (0.118 nm) is larger than that of La^{3+} ions (0.103 nm).

Fig. 3 shows the microstructures of $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics following sintering for 4 h at different temperatures. The microstructures of $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics when La^{3+} ions were replaced with different amounts of Sr^{2+} substitution indicated that the average grain size increased with the extent of Sr^{2+} substitution. The average grain size of the $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics increased from 1.2 to $1.4\ \mu\text{m}$ as x increased from 0.01 to 0.05 when the specimens were sintered at 1550°C . However, the average grain size did not considerably affect on the dielectric properties of $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics at microwave frequencies. The effect of grain size on the dielectric constant and dielectric loss of dense samples can be neglected [19]. The $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics were not dense, and grains did not grow after sintering at 1450°C for 4 h, potentially degrading the microwave dielectric properties of the $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics. Comparing the microstructures of $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics that were sintered under different temperatures indicated that the average grain size

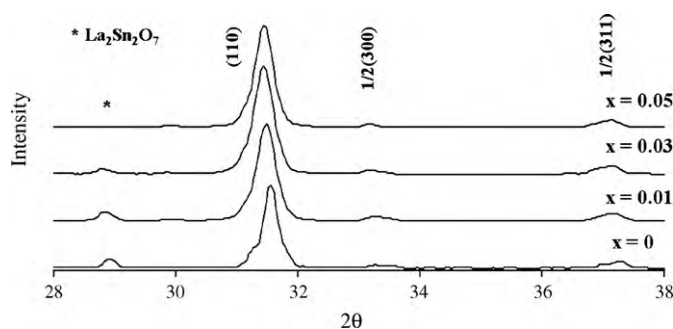


Fig. 2. X-ray diffraction patterns of $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ specimens following sintering at 1550°C for 4 h.

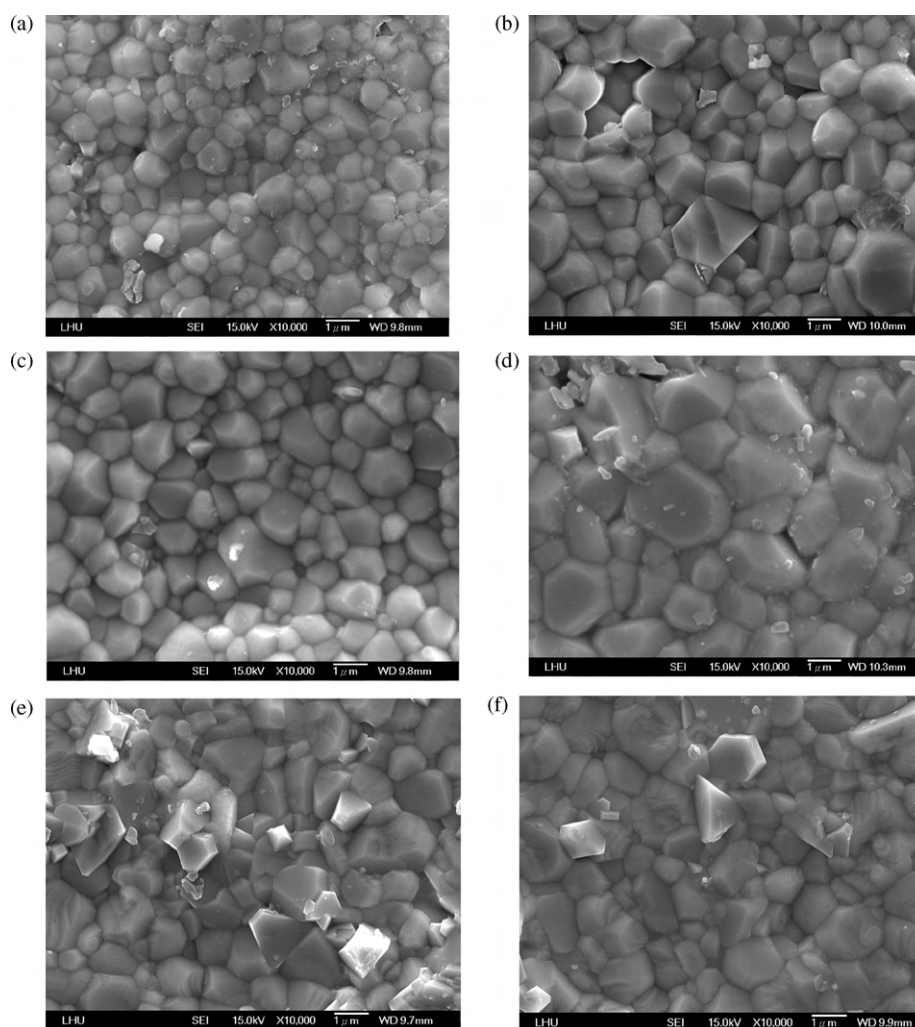


Fig. 3. Microstructures of $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics sintered under different temperatures for 4 h: (a) $x=0.01/1450^\circ\text{C}$, (b) $x=0.01/1500^\circ\text{C}$, (c) $x=0.01/1550^\circ\text{C}$, (d) $x=0.01/1600^\circ\text{C}$, (e) $x=0.03/1550^\circ\text{C}$, (f) $x=0.05/1550^\circ\text{C}$.

increased with the sintering temperature. The average grain size of the $\text{La}_{2.98/3}\text{Sr}_{0.01}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics increased from 0.9 to 1.9 μm as the temperature of sintering for 4 h increased from 1450 to 1600 $^\circ\text{C}$. The pores of $\text{La}_{2.98/3}\text{Sr}_{0.01}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics almost disappeared upon sintering at 1550 $^\circ\text{C}$ for 4 h. To identify the composition of the second phase, energy-disperse spectroscopy (EDS) analysis was carried out on the grains of $\text{La}_{2.98/3}\text{Sr}_{0.01}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics that were sintered at 1550 $^\circ\text{C}$ for 4 h, as shown in Fig. 4. According to the quantitative analysis, as shown in Table 1, the A, B, and C grains are evidently $\text{La}_{2.98/3}\text{Sr}_{0.01}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ and the D grain is $\text{La}_2\text{Sn}_2\text{O}_7$.

Fig. 5 shows the apparent densities of the $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics with different degrees of Sr^{2+} substitution, following sintering at 1450–1600 $^\circ\text{C}$ for 4 h.

The theoretical density of $\text{La}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramic is 6.63 g/cm^3 . The apparent density of the $\text{La}_{2.98/3}\text{Sr}_{0.01}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics that were sintered at 1450–1600 $^\circ\text{C}$ for 4 h was highest

when sintering was carried out at 1550 $^\circ\text{C}$, beyond which temperature, it decreased. However, the maximum apparent density decreased from 6.56 to 6.54 g/cm^3 as x increased from 0.01 to 0.05. The increase in apparent density may be caused by the decrease in the number of pores and a decrease in apparent density may

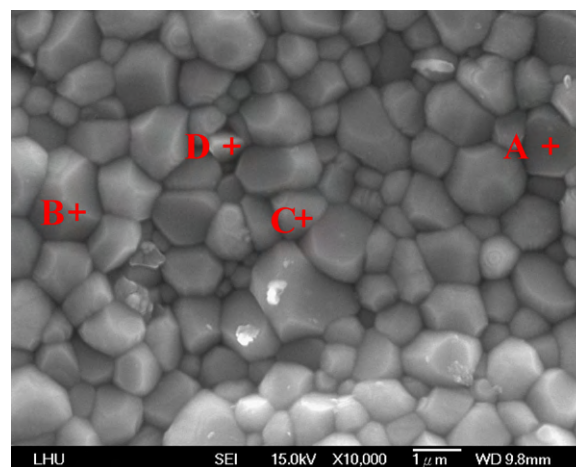


Fig. 4. Energy-disperse spectroscopy (EDS) analysis of grains of $\text{La}_{2.98/3}\text{Sr}_{0.01}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics sintered at 1550 $^\circ\text{C}$ for 4 h.

Table 1
EDS data of grains of $\text{La}_{2.98/3}\text{Sr}_{0.01}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics sintered at 1550 $^\circ\text{C}$ for 4 h.

Atomic element	La (%)	Sr (%)	Mg (%)	Sn (%)	O (%)
A	12.02	0.36	7.30	6.18	74.14
B	13.28	0.33	7.80	7.15	71.44
C	13.47	0.33	7.33	6.99	71.87
D	14.97	0	0	11.02	74.01

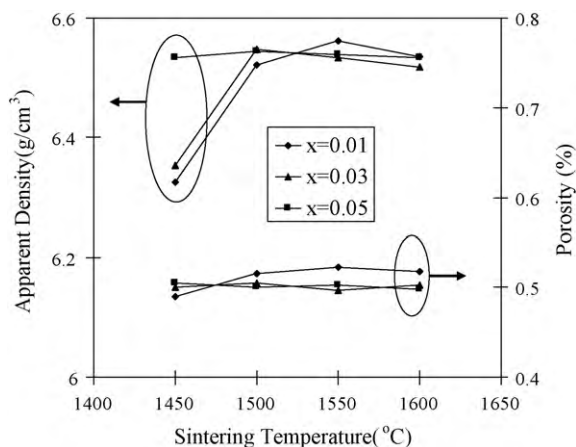


Fig. 5. Apparent densities and porosities of $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics with different degrees of Sr^{2+} substitution, following sintering at different temperatures for 4 h.

be caused by the abnormal grain growth, as shown in Fig. 3. $\text{La}_{2.98/3}\text{Sr}_{0.01}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics that were sintered at 1550°C for 4 h had a maximum apparent density of 6.56 g/cm^3 . Fig. 5 also shows the porosity of $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics with different degrees of Sr^{2+} substitution sintered at 1450 – 1600°C for 4 h. The porosity of $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics were ranged from 0.4% to 0.6% under different sintering conditions.

Fig. 6 shows the dielectric constants of $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics with different degrees of Sr^{2+} substitution, following sintering at 1450 – 1600°C for 4 h. $\text{La}_{2.98/3}\text{Sr}_{0.01}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics that were sintered at 1550°C for 4 h had a maximum dielectric constant of 20.0. The dielectric constant decreased from 20.0 to 19.9 as the x increased from 0.01 to 0.05 when the $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics were sintered at 1550°C for 4 h. The resonant frequency was approximately at 11.8 GHz. A high sintering temperature was not necessary for obtaining a high dielectric constant. The decrease in dielectric constant was associated with low densities of the ceramics. A higher density is associated with lower porosity, and, therefore, a higher dielectric constant. Table 2 shows the dielectric constant of $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics with different amounts of Sr^{2+} substitution sintered at 1500°C for 4 h. The dielectric constant of $\text{La}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$, $\text{La}_{2.98/3}\text{Sr}_{0.01}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$, $\text{La}_{2.94/3}\text{Sr}_{0.03}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$, and $\text{La}_{2.9/3}\text{Sr}_{0.05}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$, ceramics sintered at 1500°C for 4 h was 15.6, 19.8, 19.9, and 19.9,

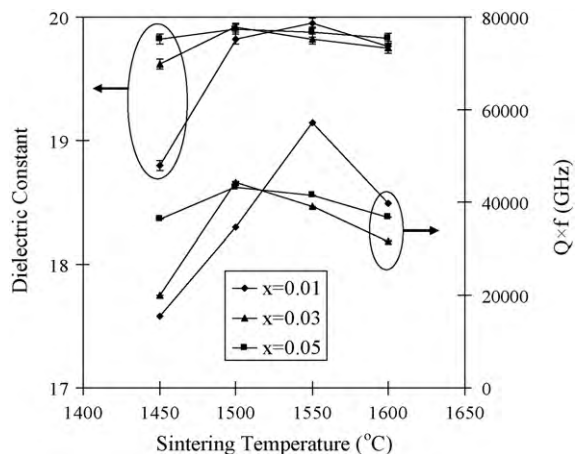


Fig. 6. Dielectric constants and $Q \times f$ of $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics with different degrees of Sr^{2+} substitution, following sintering at different temperatures for 4 h.

Table 2

Comparison the microwave dielectric properties of $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ with different amounts of Sr^{2+} substitution sintered at 1500°C for 4 h.

Material	ϵ_r	$Q \times f$ (GHz)
$\text{La}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ [16]	15.6	30,600
$\text{La}_{2.98/3}\text{Sr}_{0.01}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$	19.8	34,600
$\text{La}_{2.94/3}\text{Sr}_{0.03}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$	19.9	44,400
$\text{La}_{2.9/3}\text{Sr}_{0.05}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$	19.9	43,200

respectively. These results suggest that the dielectric constant was effectively increased by substituting some of the La^{3+} ions for Sr^{2+} ions. This fact might be explained by the volume of cation sites, as suggested by Tohdo et al. [15]. The Sr^{2+} ions, with a large ionic radius, occupy the sites of La^{3+} ions, and as the volume of cation sites increases, the dielectric constant increases.

Fig. 6 shows the $Q \times f$ of the $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics with different degrees of Sr^{2+} substitution, following sintering at 1450 – 1600°C for 4 h. The $Q \times f$ of the $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics that were sintered at 1450 – 1600°C for 4 h was highest when sintering was carried out at 1550 , 1500 , and 1500°C , beyond which temperature, it decreased, for $\text{La}_{2.98/3}\text{Sr}_{0.01}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$, $\text{La}_{2.94/3}\text{Sr}_{0.03}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$, and $\text{La}_{2.9/3}\text{Sr}_{0.05}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics, respectively. The highest $Q \times f$ of 57,100 GHz was obtained for $\text{La}_{2.98/3}\text{Sr}_{0.01}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics that were sintered at 1550°C for 4 h. The $Q \times f$ of the $\text{La}_{2.98/3}\text{Sr}_{0.01}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics increased from $15,500$ to $51,700$ GHz as the temperature of sintering for 4 h increased from 1450 to 1550°C . The relationship between the $Q \times f$ and the sintering temperature was consistent with that between the apparent density and the sintering temperature, because the microwave dielectric loss is affected by many factors, which is composed of intrinsic and extrinsic losses. Intrinsic loss is associated with the vibrational modes of the lattice. Extrinsic loss is associated with the density, porosity, second phases, impurities, oxygen vacancies, grain size, and lattice defects [20–21]. Since the $Q \times f$ of $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics was consistent with the variation of the apparent density, it is suggested to be dominated by the apparent density. Table 2 shows the $Q \times f$ of $\text{La}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$, $\text{La}_{2.98/3}\text{Sr}_{0.01}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$, $\text{La}_{2.94/3}\text{Sr}_{0.03}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$, and $\text{La}_{2.9/3}\text{Sr}_{0.05}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics sintered at 1500°C for 4 h are 30,600, 34,600, 44,400, and 43,200 GHz, respectively. These results suggest that the $Q \times f$ increased evidently by partially substituting La^{3+} ions with Sr^{2+} ions.

Fig. 7 shows the temperature coefficient of resonant frequency (τ_f) of $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics with different degrees of Sr^{2+} substitution, following sintering at 1450 – 1600°C for 4 h. Generally, τ_f is related to the composition, the amounts of additive, and the second phases that are presented in the

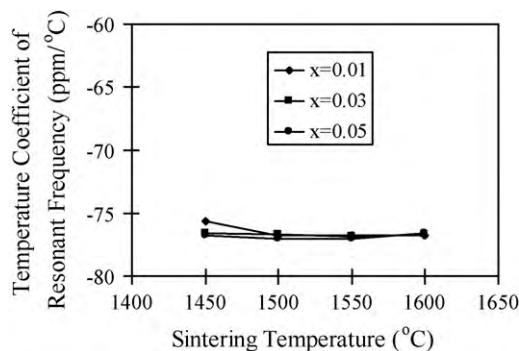


Fig. 7. τ_f of $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics with different degrees of Sr^{2+} substitution, following sintering at different temperatures for 4 h.

ceramics [14]. Although a second phase $\text{La}_2\text{Sn}_2\text{O}_7$ was found, small amounts of second phase $\text{La}_2\text{Sn}_2\text{O}_7$, which influence the τ_f of $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics, were small. No significant variation in τ_f of $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics with sintering temperature over the entire range of sintering temperatures considered herein was observed. Since the composition of $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics with a fixed amount of Sr^{2+} substitution did not vary with sintering temperature. The τ_f of $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics were measured as a function of the amounts of Sr^{2+} substitution in this experiment. A τ_f of $-77 \text{ ppm}/^\circ\text{C}$ was measured for $\text{La}_{2.98/3}\text{Sr}_{0.01}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics that were sintered at 1550°C for 4 h.

4. Conclusions

The effects of the degree of Sr^{2+} substitution and sintering temperature on the microwave dielectric properties of $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics were studied. The microwave dielectric properties of $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics were improved by substituting La^{3+} ions with Sr^{2+} ions. The diffraction peaks of $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics shifted to lower angles as x increased from 0 to 0.05. The X-ray diffraction peaks of $\text{La}_{2.98/3}\text{Sr}_{0.01}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramic did not vary significantly with sintering temperatures. $\text{La}_{2.98/3}\text{Sr}_{0.01}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics that were sintered at 1550°C for 4 h had an apparent density of 6.56 g/cm^3 , a dielectric constant of 20.0, a $Q \times f$ of 57,100 GHz, and a temperature coefficient of resonant frequency (τ_f) of $-77 \text{ ppm}/^\circ\text{C}$. The microwave dielectric properties of $\text{La}_{(1-2x/3)}\text{Sr}_x(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics depend strongly on the apparent density and microstructures.

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