# *Effect of Sintering Time on Microwave Dielectric Properties of $La_2Sn_2O_7$ Ceramics*

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*Abstract - The microwave dielectric properties of $La_2Sn_2O_7$ ceramics were investigated with a view to their application in mobile communication. The $La_2Sn_2O_7$ ceramics were prepared by the conventional solid-state method with various sintering times. A maximum apparent density of 6.60 g/cm$^3$ was obtained for $La_2Sn_2O_7$ ceramic, sintered at 1550 $^O$C for 34 h. Dielectric constants ($\varepsilon_r$) of 11.9-14.3 and quality factor ($Q \times f$) of 28,100-38,300 GHz were obtained at sintering times in the range 33-35 h at a sintering temperature of 1550 $^O$C. A dielectric constant ($\varepsilon_r$) of 14.3, a quality factor ($Q \times f$) of 38,300 GHz, and a temperature coefficient of resonant frequency ($\tau_f$) of -53 ppm/ $^O$C were obtained when $La_2Sn_2O_7$ ceramics were sintered at 1550 $^O$C for 34h.*

*Keywords - $La_2Sn_2O_7$, Dielectric constant, Quality factor, Temperature coefficient of resonant frequency.*

## *I. INTRODUCTION*

Materials that are to be used in microwave devices must have three dielectric characteristics - a high dielectric constant, a high quality factor, and a near-zero temperature coefficient of resonant frequency. These enable small devices with low loss and high temperature stability, respectively, to be fabricated [1-2]. The benefits of using complex perovskite ceramics $A(B'_{0.5}B''_{0.5})O_3$ (A=$Me^{2+}$, $Me^{3+}$; B'=$Me^{2+}$, $Me^{3+}$; B"=$Me^{4+}$, $Me^{5+}$, $Me^{6+}$) are reportedly associated with their excellent dielectric properties at microwave frequencies [3-9]. Numerous investigations of researches on the $La(Mg_{0.5}Sn_{0.5})O_3$ ceramics and related ceramic systems have explored their potential application in resonators, filters and antennas in the modern communication systems, including radar and global positioning systems (GPS), which are operated at microwave frequencies. A dielectric constant of 19.7, a $Q\times f$ of 43,300 GHz and a $\tau_f$ of -83 ppm/$^O$C was obtained for $La(Mg_{0.5}Sn_{0.5})O_3$ ceramics with 0.5 wt% CuO additive, sintered at 1550 $^O$C for 4 h [10]. A dielectric constant of 19.7, a $Q\times f$ of 45,000 GHz and a $\tau_f$ of -85 ppm/$^O$C was obtained for $La(Mg_{0.5}Sn_{0.5})O_3$ ceramics with 0.5 wt% $B_2O_3$ additive sintered at 1500 $^O$C for 4h [11]. A dielectric constant of 20.3, a $Q\times f$ of 48,400 GHz, and a $\tau_f$ of −87.5 ppm/$^O$C were obtained for $La(Mg_{0.5}Sn_{0.5})O_3$ ceramics with 0.5 wt% $V_2O_5$ additive sintered at 1500 $^O$C for 4 h [12]. $La(Mg_{0.5}Sn_{0.5})O_3$ was the main crystalline phase, which was accompanied by small amounts of $La_2Sn_2O_7$ as the second phase [10-12]. The formation of the Sn-rich second phase $La_2Sn_2O_7$ was caused by the loss of MgO upon ignition. The formation of the second phase, $La_2Sn_2O_7$, affected the microwave dielectric properties of $La(Mg_{0.5}Sn_{0.5})O_3$ ceramics. However, no technical information on the microwave dielectric properties of $La_2Sn_2O_7$ ceramics is available in the published literature. This fact motivates this investigation of the microwave dielectric properties of $La_2Sn_2O_7$ ceramics.

$La_2Sn_2O_7$ ceramics were synthesized herein using the conventional mixed-oxide method. The effects of the sintering time on the microwave dielectric properties of $La_2Sn_2O_7$ ceramics were explored. The dielectric properties of the $La_2Sn_2O_7$ ceramics at microwave frequencies were found to vary with the sintering time. For further understanding of these different microwave dielectric properties, they were analyzed by densification, X-ray diffraction (XRD), and microstructural observations.

## *II. EXPERIMENTAL PORCEDURE*

The starting raw chemicals were highly pure $La_2O_3$ and $SnO_2$ powders (99.9%). The prepared compound was $La_2Sn_2O_7$. Specimens were prepared by the conventional mixed-oxide method. The raw material was weighed out in stoichiometric proportions, ball-milled in alcohol, dried, and then calcined at 1200 $^O$C for 4 h. The calcined powder was re-milled for 12 h using PVA solution as a binder. The obtained powder was then crushed into a fine 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 1550 $^O$C for 33-35 h in air. Both the heating rate and the cooling rate were set to 10 $^O$C/min.

Following sintering, the phases of the samples were investigated by X-ray diffraction. An X-ray Rigaku D/MAX-2200 data was used with Cu$K\alpha$ radiation (at 30 kV and 20 mA) and a graphite monochromator in the 2θ range of 10$^o$-80$^o$. Scanning electron microscopy (SEM; JEOL JSM-6500F) was utilized to elucidate the microstructures of the specimens. The apparent densities of which were measured using the Archimedes method with distilled water as the liquid. The microwave dielectric properties of the specimens were measured using the postresonator method of developed by Hakki and Coleman [13]. The postresonator scheme adopted 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 approximately 1.6, which is in the range permitted by Kobayashi and Katoh [14]. 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 N5230A network analyzer. The resonance characteristics depended on the size and dielectric properties of the specimen. The microwave energy was coupled using electric-field probes. The $TE_{011}$ resonant mode was optimal for determining the dielectric constant, and the loss factor of the specimen. The Agilent N5230A 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 scheme for measuring $\tau_f$ 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 $^{O}$C. The $\tau_f$ value (ppm/$^{O}$C) can be determined by noting the change in resonant frequency,

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

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

### *III. EXPERIMENTAL PORCEDURE*

Figure 1 displays the X-ray diffraction patterns of $La_2Sn_2O_7$ ceramics sintered at 1550 $^{O}$C for 33-35 h. Clearly, $La_2Sn_2O_7$ is the main crystalline phase. As evidenced by Fig.1, the spectral angles of the X-ray diffraction peaks were the same following sintering at 1550 $^{O}$C for 33-35 h. All of the peaks were indexed according to the cubic unit cell.

Figure 2 shows the microstructures of $La_2Sn_2O_7$ ceramics under various sintering conditions. The $La_2Sn_2O_7$ ceramics was not dense, and grains did not grow following sintering at 1550 $^{O}$C for 33 h. As the sintering time was prolonged from 33 to 34 h, the holes were eliminated and densification occurred. The holes of $La_2Sn_2O_7$ ceramics almost disappeared upon sintering at 1550 $^{O}$C for 34 h. The holes may influence the microwave dielectric properties of the $La_2Sn_2O_7$ ceramics. To identify the compositions of the second phase, an energy-disperse spectroscopy (EDS) analysis of the grains of $La_2Sn_2O_7$ ceramics that were sintered at 1500 $^{O}$C for 34 h was carried out, as shown in Fig. 2. According to the quantitative analysis, displayed in Table 1, the A and B grains are $La_2Sn_2O_7$.

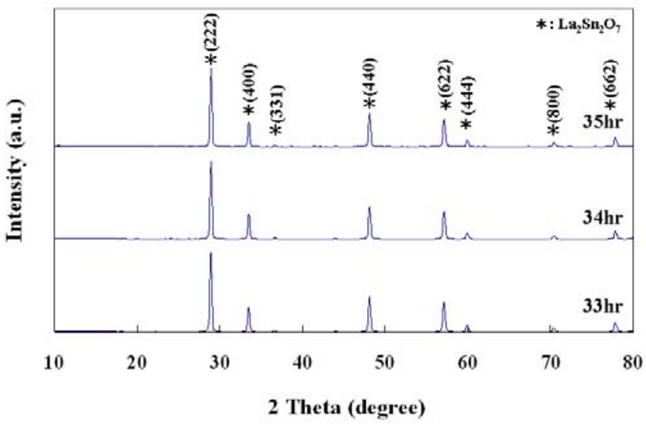
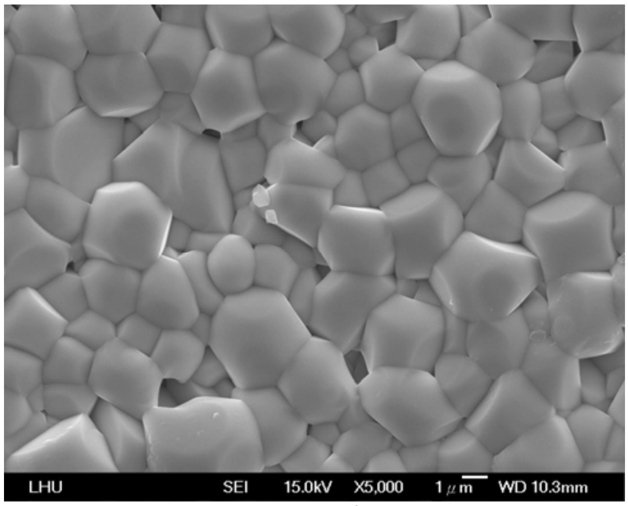
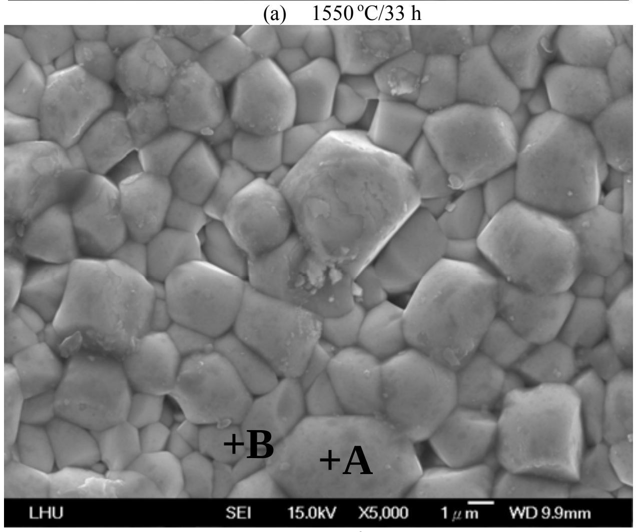


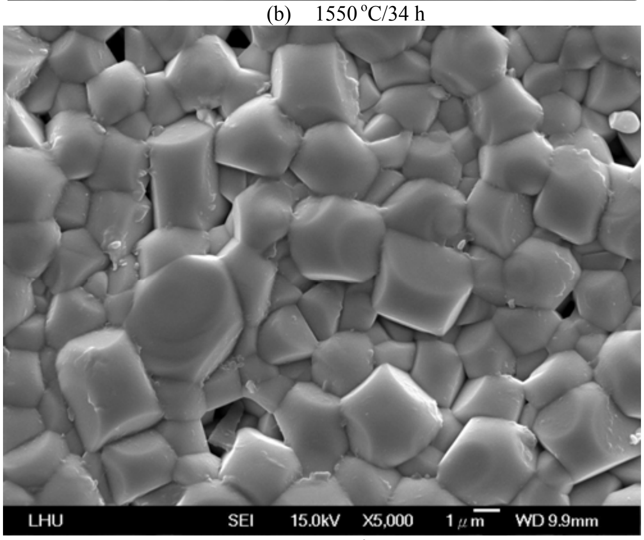

Fig. 1 X-ray diffraction patterns of $La_2Sn_2O_7$ ceramics sintered at 1550 $^{O}$C for 33-35 h.




(a) 1550 °C/33 h




(b) 1550 °C/34 h




(c) 1550 °C/35 h

Fig. 2 Microstructures of $La_2Sn_2O_7$ ceramics sintered under various sintering conditions.

TABLE I
DATA CONCERNING GRAINS OF $La_2Sn_2O_7$ CERAMICS SINTERED AT 1550 $^{O}$C FOR 34 H.

| Atomic element | Grains | |
|---|---|---|
| | A | B |
| La | 11.46 | 16.95 |
| Sn | 12.22 | 15.21 |
| O | 76.31 | 67.84 |

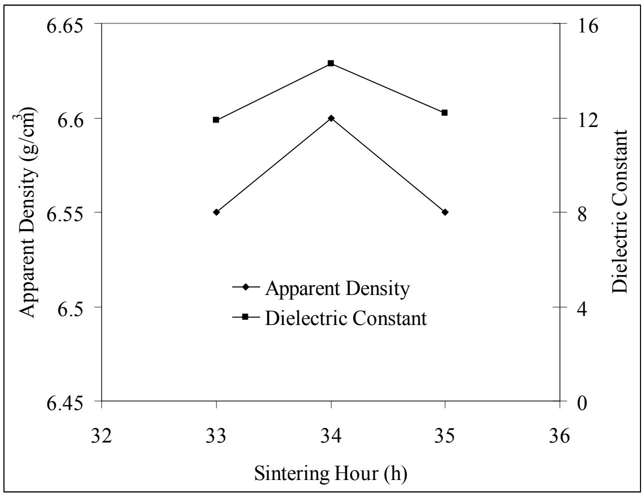


Fig. 3 Apparent densities and dielectric constant of $La_2Sn_2O_7$ ceramics sintered at 1550 $^OC$ for 33-35 h.

Figure 3 displays the apparent densities and dielectric constants of the $La_2Sn_2O_7$ ceramics that were sintered at 1550 $^OC$ for 33-35 h. The apparent densities of the $La_2Sn_2O_7$ ceramics sintered at 1550 $^OC$ increased with sintering time to a maximum at 34 h, and thereafter decreased. $La_2Sn_2O_7$ ceramics that were sintered at 1550 $^OC$ for 34 h had a maximum apparent density of 6.6 g/cm$^3$. The increase in apparent density may be caused by the decrease in the number of pores as presented in Fig. 2. The relationship between the dielectric constant and the sintering conditions was similar to that between the apparent density and the sintering conditions. The dielectric constant of $La_2Sn_2O_7$ ceramics that were sintered at 1550 $^OC$ ranged from 11.9 to 14.3 as the sintering time varied from 33 to 35 h. The $La_2Sn_2O_7$ ceramics that were sintered at 1550 $^OC$ for 34 h had the highest dielectric constant of 14.3. The decrease in dielectric constant was associated with the fall in apparent density of the ceramics. A higher apparent density is associated with a lower porosity and, therefore, a higher dielectric constant.

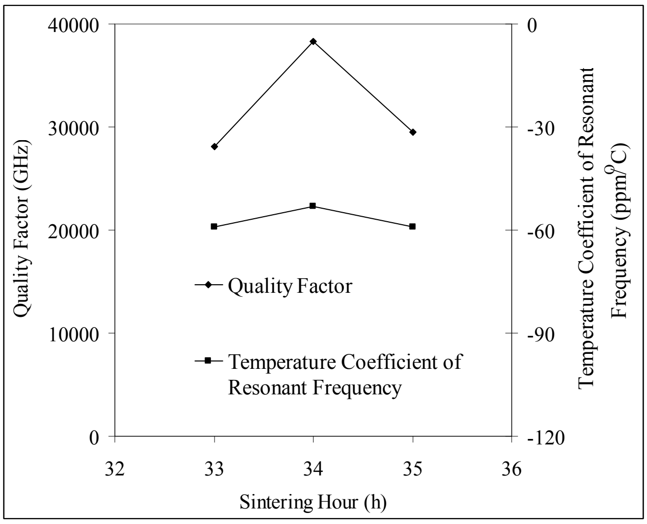


Fig. 4 Quality factor and temperature coefficient of resonant frequency of $La_2Sn_2O_7$ ceramics sintered at 1550 $^OC$ for 33-35 h.

Figure 4 presents the $Q\times f$ and temperature coefficient of resonant frequency ($\tau_f$) of the $La_2Sn_2O_7$ ceramics that were sintered at 1550 $^OC$ for 33-35 h, respectively. The $Q\times f$ of the $La_2Sn_2O_7$ ceramics that were sintered at 1550 $^OC$ increased from 28,100 to 38,300 GHz as the sintering time increased from 33 to 34 h. The highest $Q\times f$ of 38,300 GHz was obtained for $La_2Sn_2O_7$ ceramics that were sintered at 1550 $^OC$ for 34 h. The relationship between the Q×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 several factors, and consists of intrinsic and extrinsic components. Intrinsic loss is associated with the lattice vibrational modes. Extrinsic loss is related to the density, porosity, second phases, impurities, oxygen vacancies, grain size and lattice defects [15-16]. Since the $Q\times f$ of $La_2Sn_2O_7$ ceramics was consistent with the variation of the apparent density, the $Q\times f$ of $La_2Sn_2O_7$ ceramics is suggested to be dominated by the apparent density. Generally, $\tau_f$ is related to the composition, the amount of additive, and the second phases that are present in ceramics. Since the composition of $La_2Sn_2O_7$ ceramics did not vary with sintering time, no significant variation in $\tau_f$ with sintering time over the entire range of these variables was observed. The $\tau_f$ of $La_2Sn_2O_7$ ceramics ranged from -59 to -53 ppm/$^OC$ as the sintering conditions varied. A $\tau_f$ of -53 ppm/$^OC$ was measured for $La_2Sn_2O_7$ ceramic that was sintered at 1550 $^OC$ for 34 h.

## *IV. CONCLUSIONS*

The effects of sintering time on the microwave dielectric properties of $La_2Sn_2O_7$ ceramics were examined. The X-ray diffraction patterns of the $La_2Sn_2O_7$ ceramics do not significantly vary with sintering time. $La_2Sn_2O_7$ ceramics that were sintered at 1550 $^OC$ for 4 h had an apparent density of 6.6 g/cm$^3$, a dielectric constant of 14.3, and a $Q\times f$ of 38,300 GHz. The dielectric constant and $Q\times f$ of $La_2Sn_2O_7$ ceramics depend strongly on the apparent density. $La_2Sn_2O_7$ ceramics that were sintered at 1550 $^OC$ for 34 h had a temperature coefficient of resonant frequency ($\tau_f$) of -53 ppm/$^OC$.

## *ACKNOWLEDGMENT*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