

Improved microwave dielectric properties of $\text{Nd}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics by substituting Mg^{2+} with Zn^{2+}

Yih-Chien Chen^{*}, Kuang-Chiung Chang, Shi-Li Yao

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

Received 18 December 2011; received in revised form 2 March 2012; accepted 19 March 2012

Available online 28 March 2012

Abstract

This study elucidates the microwave dielectric properties and microstructures of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics for their potential applications in microwave devices. The $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics were prepared by the conventional solid-state method with various sintering temperatures. X-ray diffraction (XRD) patterns of the $\text{Nd}(\text{Mg}_{0.4}\text{Zn}_{0.1}\text{Sn}_{0.5})\text{O}_3$ ceramics revealed only a slight variation of phase with sintering temperatures. Additionally, a dielectric constant (ϵ_r) of 19.5, a quality factor ($Q \times f$) of 129,200 GHz, and a temperature coefficient of resonant frequency (τ_f) of $-66 \text{ ppm}/^\circ\text{C}$ were obtained for $\text{Nd}(\text{Mg}_{0.4}\text{Zn}_{0.1}\text{Sn}_{0.5})\text{O}_3$ ceramics sintered at 1500°C for 4 h.

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Keywords: $\text{Nd}(\text{Mg}_{0.4}\text{Zn}_{0.1}\text{Sn}_{0.5})\text{O}_3$; Dielectric constant; Quality factor; Temperature coefficient of resonant frequency

1. Introduction

Of the many commercial applications using microwave devices include mobile radio and wireless communications. However, microwave devices are limited in size, bandwidth, and efficiency, all imposed by the dielectric substrate. Three microwave dielectric properties of dielectric substrates must be considered for microwave devices, i.e. a high dielectric constant, high quality factor and near-zero temperature coefficient of resonant frequency [1–5]. High dielectric constant and a near zero temperature coefficient of resonant frequency are essential to ensure a compact size and high temperature stability, respectively. Quality factor is a representative of the loss of microwave devices. Many investigations of $\text{Nd}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics and related ceramic systems have investigated their potential application in resonators, filters and antennas in modern communication systems, e.g. radar and wireless local area network (WLAN), which operate at microwave frequencies. A dielectric constant of 19.3, quality factor ($Q \times f$) of 43,300 GHz, and temperature

coefficient of resonant frequency of $-57 \text{ ppm}/^\circ\text{C}$ were obtained when $\text{Nd}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics sintered at 1550°C for 4 h [6]. A dielectric constant around 18.9 and a $Q \times f$ of 32,300 GHz were also obtained for 0.25 wt% B_2O_3 -doped $\text{Nd}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics sintered at 1500°C for 4 h [7]. Related investigations found that several solid solutions combining two compounds have a higher value of $Q \times f$ than that of both end members [8–10], subsequently, motivating this study on the role of replacing Mg^{2+} (0.072 nm) by Zn^{2+} (0.074 nm) in forming $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics [11]. In this study, $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics are synthesized and some of the Mg^{2+} ions are replaced by Zn^{2+} ions to enhance their dielectric constant and $Q \times f$. Additionally, exactly how sintering temperature affects the microwave dielectric properties of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics is examined. $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics are synthesized using the conventional mixed-oxide method with experimental results demonstrating that they have a higher dielectric constant and $Q \times f$ than that of $\text{Nd}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics. Moreover, the microwave dielectric properties of the $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics vary with the extent of Zn^{2+} substitution and sintering temperatures. These microwave dielectric properties are analyzed by densification, X-ray diffraction (XRD) patterns, along with an observation of their microstructures.

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

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

2. Experimental procedure

The starting raw chemicals were Nd_2O_3 (99.99%), MgO (98.0%), ZnO (99.9%), and SnO_2 (99.0%) powders. The prepared composition was $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$. Specimens were prepared using the conventional mixed-oxide method. The raw material was weighed in stoichiometric proportions, 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 fine powder was then crushed into a finer powder through a sieve with a 200 mesh. Next, this finer powder was pressed axially at 2000 kg/cm^2 into pellets with a diameter of 11 mm and a thickness of 6 mm. The specimens subsequently obtained were sintered at temperatures ranging from 1350 to 1600°C for 4 h in air. Both the heating rate and the cooling rate were set to 10°C/min .

Following sintering, phases of the samples were examined by X-ray diffraction. An X-ray Rigaku D/MAX-2200 was utilized with $\text{Cu K}\alpha$ radiation (at 30 kV and 20 mA) and a graphite monochromator in the 2θ range of 10 – 70° . Microstructures of the specimens were then examined using scanning electron microscopy (SEM; JEOL JSM-6500F) and energy dispersive X-ray spectrometer (EDS). The grain sizes were determined by the line intercept method. The apparent densities of the specimens were measured using the liquid Archimedes method, along with distilled water. Next, the microwave dielectric properties of the specimens were measured by the postresonator method developed by Hakki and Coleman [12]. This method utilizes a specimen in the form of a cylinder of diameter D and length L . The specimens whose microwave dielectric properties were measured had an aspect ratio D/L of approximately 1.6, which is in the permitted range as determined by Kobayashi and Katoh [13]. The cylindrical resonator was sandwiched between two conducting plates. Two small antennas were positioned near 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. Notably, the resonance characteristics depended on the size and microwave dielectric properties of the specimen. Next, the microwave energy was coupled using electric-field probes. The TE_{011} resonant mode was most effective in determining the dielectric constant and the loss factor of the specimen. The Agilent E5071C network analyzer was adopted to identify the TE_{011} resonant frequency of the dielectric resonator, and the dielectric constant and quality factor were calculated. The approach for measuring τ_f was the same as that for measuring the dielectric constant. The test cavity was placed in a chamber and, then, the temperature was increased from 25 to 75°C . The τ_f value ($\text{ppm}/^\circ\text{C}$) was determined according to 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 refer to the resonant frequencies at T_1 and T_2 , respectively.

3. Results and discussion

Fig. 1 presents the X-ray diffraction patterns of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics sintered under different temperatures for 4 h. Obviously, $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ is

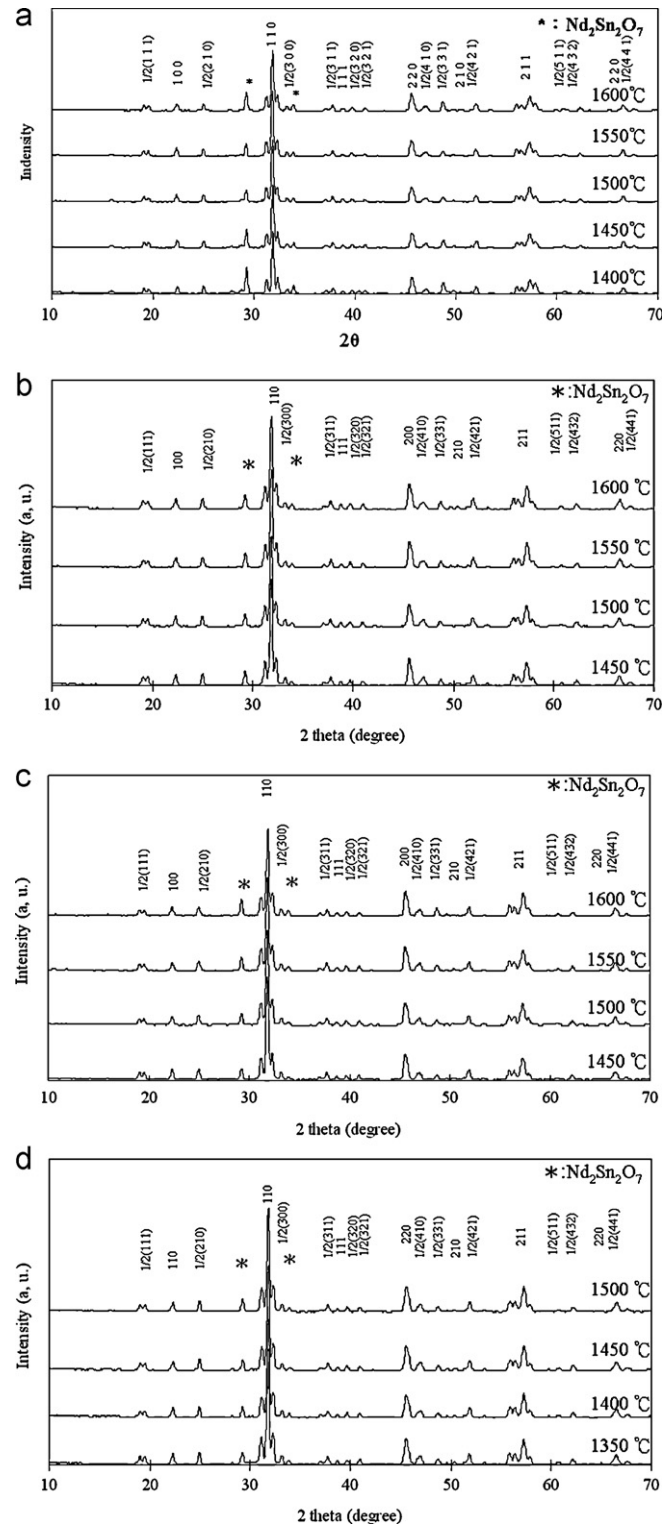


Fig. 1. X-ray diffraction patterns of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics sintered under different temperatures for 4 h: (a) $x = 0$, (b) $x = 0.05$, (c) $x = 0.10$, (d) $x = 0.15$.

Table 1

Optimum sintering temperature, tolerance factor, and grain size of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics.

x	Optimum sintering temperature ($^{\circ}\text{C}$)	Tolerance factor	Grain size (μm)
0	1550	0.8970	1.33
0.05	1550	0.8966	1.57
0.10	1500	0.8962	1.59
0.15	1400	0.8958	1.31

the main crystalline phase, accompanied by small amounts of $\text{Nd}_2\text{Sn}_2\text{O}_7$ as the second phase. $\text{Nd}_2\text{Sn}_2\text{O}_7$ with a cubic crystal structure (ICDD-PDF #88-0448) was identified and eliminating it completely from the sample prepared by mixed oxide route was relatively difficult. According to Fig. 1, the (2 2 2) and (4 0 0) peaks of the $\text{Nd}_2\text{Sn}_2\text{O}_7$ ceramics appeared at 29.253° and 33.905° , respectively. Also, a series of extra peaks correlated with superlattice reflections. As is well known, unit-cell doubling originates from two mechanisms, i.e. cation ordering and systematic tilting of oxygen octahedral. The cation ordering of Mg^{2+} and Sn^{4+} in B-sites of $\text{Nd}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ led to a doubling of the unit cell. Owing to the presence of $\{h + 1/2, k + 1/2, l + 1/2\}$ type, superlattice reflections were indexed using half integer Miller indices. For

systematic tilting of oxygen octahedral is associated with the rotation of oxygen octahedral without distorting it. According to Glazer, superlattice reflections, with specific combinations of odd (o) and even (e) Miller indices, reveal particular deviations of the structure from an undistorted cubic structure, including octahedral in-phase tilting (ooe, oeo, eoo), anti-phase tilting (ooo, $h + k + l > 3$), and anti-parallel displacement of A-cations (eoo, eoe, oee) [14]. The (1/2(2 1 0), 1/2(3 0 0), 1/2(3 2 0), 1/2(4 1 0), 1/2(4 2 1), 1/2(4 3 2), and 1/2(4 4 1)) additional peaks demonstrate A-site cation displacement, in which (1/2(3 1 1), 1/2(3 3 1), and 1/2(5 1 1)) peaks indicate anti-phase tilting, while the 1/2(3 2 1) peaks denote in-phase tilting. Notably, the 1/2(1 1 1) provides evidence of 1:1 B-site cation ordering. Also, the X-ray diffraction patterns of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics only slightly vary with sintering temperature. The tolerance factors of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics decrease from 0.8970 to 0.8958 as x was increased from 0 to 0.15, as shown in Table 1. The tolerance factor was calculated using the ionic radius data of Shannon [11]. The tolerance factors of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ series belong to the anti-phase and in-phase tilted region [15], which is in agreement with those of X-ray diffraction patterns. The perovskite cell deformed and its symmetry lowered from cubic symmetry when tolerance factor deviated from one. Deviation from cubic symmetry resulted in

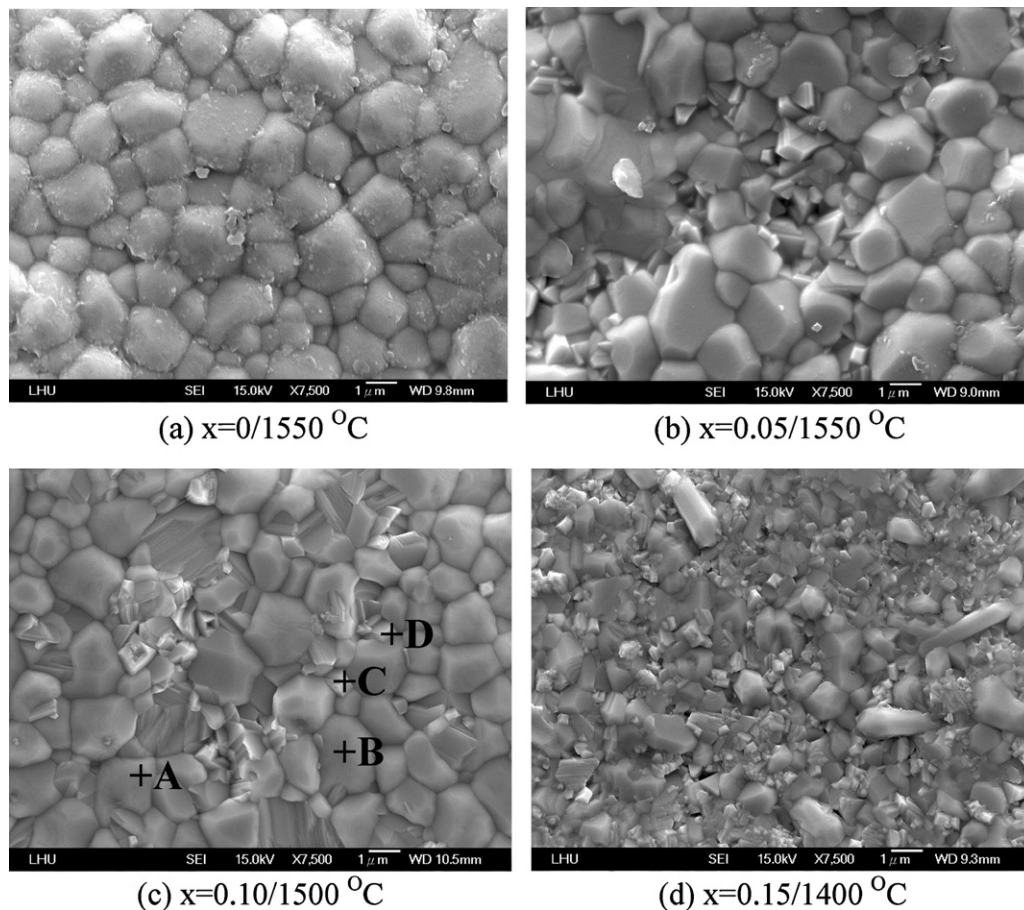


Fig. 2. Microstructures of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics sintered under different temperatures for 4 h: (a) $x = 0/1550^{\circ}\text{C}$, (b) $x = 0.05/1550^{\circ}\text{C}$, (c) $x = 0.10/1500^{\circ}\text{C}$, and (d) $x = 0.15/1400^{\circ}\text{C}$.

extra polarizations, which is reflected in the dielectric constant, ultimately resulting in a higher dielectric constant [16]. The second phase was conducted using Rietveld refinement. The amount of main phase of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics sintered at optimum sintering temperature for 4 h increased from 92.59% to 92.96% when increasing x from 0 to 0.15. Moreover, the formation of second phase of $\text{Nd}_2\text{Sn}_2\text{O}_7$ affected the apparent density and microwave dielectric properties of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics.

Fig. 2 shows the microstructures of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics following sintering for 4 h at optimum sintering temperature. The microstructures of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics that were sintered at optimum sintering temperature revealed that the average grain size increased from 1.33 to 1.59 μm when increasing x from 0 to 0.10, then decreased significantly from 1.59 to 1.31 μm when increasing x from 0.10 to 0.15, as shown in Table 1. Next, composition of the second phase was identified by an energy-disperse spectroscopy (EDS) analysis on the grains of $\text{Nd}(\text{Mg}_{0.4}\text{Zn}_{0.1}\text{Sn}_{0.5})\text{O}_3$ ceramics sintered at 1500 °C for 4 h, as shown in Fig. 2(c). Quantitative analysis revealed that A and B grains are evidently $\text{Nd}(\text{Mg}_{0.4}\text{Zn}_{0.1}\text{Sn}_{0.5})\text{O}_3$ and the C and D grains are $\text{Nd}_2\text{Sn}_2\text{O}_7$.

Fig. 3 shows the apparent densities of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics with different degrees of Zn^{2+} substitution, following sintering at 1350–1600 °C for 4 h. Notably, the apparent density of the $\text{Nd}(\text{Mg}_{0.4}\text{Zn}_{0.1}\text{Sn}_{0.5})\text{O}_3$ ceramics sintered at 1450–1600 °C for 4 h was greatest when sintering was undertaken at 1500 °C, beyond which temperature, the apparent density decreased. The maximum apparent densities of the $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics sintered at 1350–1600 °C for 4 h decreased from 6.97 to 6.90 g/cm^3 as x increased from 0 to 0.05, then increased from 6.90 to 7.03 g/cm^3 as x increased from 0.05 to 0.15. Since the Zn atom has a larger mass than that of the Mg atom, the apparent density of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics is expected to increase with an increasing x . However, the apparent density of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics decreased as x increased from 0 to 0.05. The decrease in apparent density of the specimens is

associated with decrease in the amount of second phase as x increased from 0 to 0.05. The theoretical densities of $\text{Nd}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ and $\text{Nd}_2\text{Sn}_2\text{O}_7$ are 6.961 and 7.18 g/cm^3 , respectively. Table 2 shows the theoretical density of the $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics with different degrees of Zn^{2+} substitution. The theoretical density of the composites can be calculated by the following equation:

$$D_{\text{composite}} = V_1 D_1 + V_2 D_2, \quad (2)$$

where $D_{\text{composite}}$ denotes the calculated theoretical density of the composite, V_1 and V_2 represent the volume fraction of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ and $\text{Nd}_2\text{Sn}_2\text{O}_7$, respectively, and D_1 and D_2 refer to the theoretical density of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ and $\text{Nd}_2\text{Sn}_2\text{O}_7$, respectively. Table 2 lists the relative densities of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics with different degrees of Zn^{2+} substitution, followed by sintering at optimum sintering temperature. The relative densities of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics with different degrees of Zn^{2+} substitution, which were sintered at optimum sintering temperature for 4 h, exceeded 98%.

Fig. 4 shows the dielectric constants of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics with different degrees of Zn^{2+} substitution, followed by sintering at 1350–1600 °C for 4 h. $\text{Nd}(\text{Mg}_{0.35}\text{Zn}_{0.15}\text{Sn}_{0.5})\text{O}_3$ ceramics sintered at 1400 °C for 4 h had a maximum dielectric constant of 19.7. A high dielectric constant of $\text{Nd}(\text{Mg}_{0.35}\text{Zn}_{0.15}\text{Sn}_{0.5})\text{O}_3$ ceramics sintered at 1400 °C for 4 h did not depend on a high sintering temperature. The decrease in dielectric constant was associated with low apparent densities of the ceramics. As is well known, a higher density implies a lower porosity, and, therefore, a higher dielectric constant. The dielectric constants of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics were almost the same as x increased from 0 to 0.05, then increased from 19.3 to 19.7 as x increased from 0.05 to 0.15 when the $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics were sintered at optimum sintering temperature. Of the many factors affecting the dielectric constant of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics, included the ionic polarization, second phase, and relative density. For ionic polarization, Tohdo et al. [17] suggested calculating the dielectric constant by using the Clausius–Mossotti equation:

$$\epsilon_r = \frac{3V_m + 8\pi\alpha_D}{3V_m - 4\pi\alpha_D} \quad (3)$$

where V_m represents the molar volume and α_D is the sum of the ionic polarizabilities of individual ions. The dielectric constant calculated by Eq. (3) was the intrinsic factor of the dielectric property. Dielectric constants thus depend on the molar volume and ionic polarization. As indicated by Eq. (3), a smaller molar volume or a larger ionic polarization implies a larger dielectric constant. The extent to which ionic polarization influences the dielectric constant is significantly greater than molar volume does. The ionic polarizations of Zn^{2+} ion and Mg^{2+} ions are 2.04 and 1.32 \AA^3 , respectively [18,19]. The sum of ionic polarizations of individual ions of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics increased as x increased, explaining why the dielectric constant of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics is expected to

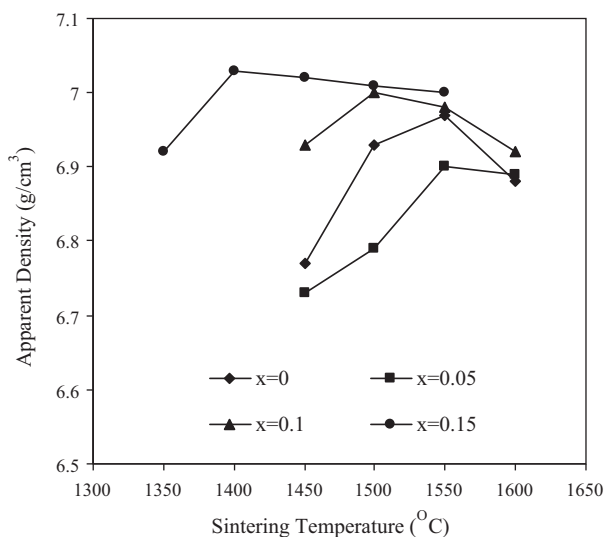


Fig. 3. Apparent densities of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics with various degrees of Zn^{2+} substitution, following sintering at various temperatures for 4 h.

Table 2

Theoretical density, relative density, dielectric constant, and quality factor of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics.

x	Theoretical density (g/cm^3)	Relative density (%)	Dielectric constant	Quality factor (GHz)
0	6.98	99.86	19.3	43,300
0.05	7.03	98.15	19.2	70,300
0.10	7.08	98.87	19.5	129,200
0.15	7.13	98.60	19.7	77,400

increase. During the second phase, a dielectric constant of 17.0 was obtained for $\text{Nd}_2\text{Sn}_2\text{O}_7$ ceramics. The dielectric constant of the composites can be calculated by the following mixture rule:

$$L\epsilon_r = V_1 L\epsilon_{r1} + V_2 L\epsilon_{r2}, \quad (4)$$

where ϵ_r denotes the dielectric constant of the composite, V_1 and V_2 represent the volume fraction of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ and $\text{Nd}_2\text{Sn}_2\text{O}_7$, respectively, and ϵ_{r1} and ϵ_{r2} refer to the dielectric constant of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ and $\text{Nd}_2\text{Sn}_2\text{O}_7$, respectively. Since the amount of second phase decreased as x increased from 0 to 0.15, we can infer that the dielectric constants of specimens increase as x increased. However, in this study, the dielectric constants of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics are almost the same as x is increased from 0 to 0.05. This finding can be explained by the relative density. Relative density is an extrinsic factor in controlling the dielectric constant. As mentioned earlier, densification plays an important role in controlling the dielectric constant. In this study, the relative density decreased from 99.86% to 98.15% as x increased from 0 to 0.05, as shown in Table 2. Moreover, the dielectric constant of $\text{Nd}(\text{Mg}_{0.35}\text{Zn}_{0.15}\text{Sn}_{0.5})\text{O}_3$ ceramics sintered at 1350 °C exceeded that of $\text{Nd}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics sintered at 1550 °C. A large sintering temperature reduction of approximately 200 °C was achieved by replacing Mg^{2+} with Zn^{2+} .

Fig. 5 shows the $Q \times f$ value of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics with different degrees of Zn^{2+} substitution, followed by sintering at 1350–1600 °C for 4 h. The relationship between the

$Q \times f$ and the sintering temperature of $\text{Nd}(\text{Mg}_{0.4}\text{Zn}_{0.1}\text{Sn}_{0.5})\text{O}_3$ ceramics was consistent with that between the apparent density and the sintering temperature, because many factors affect the microwave dielectric loss, which comprises intrinsic and extrinsic losses. While the former is associated with the vibration modes of a lattice, the latter is associated with density, porosity, second phases, impurities, oxygen vacancies, grain size, and lattice defects [20,21]. The observation that the $Q \times f$ value of $\text{Nd}(\text{Mg}_{0.4}\text{Zn}_{0.1}\text{Sn}_{0.5})\text{O}_3$ ceramics correlated with the variation of the apparent density implies that the apparent density dominates the $Q \times f$ value of $\text{Nd}(\text{Mg}_{0.4}\text{Zn}_{0.1}\text{Sn}_{0.5})\text{O}_3$ ceramics. Additionally, $\text{Nd}(\text{Mg}_{0.4}\text{Zn}_{0.1}\text{Sn}_{0.5})\text{O}_3$ ceramics sintered at 1500 °C for 4 h achieved a maximum $Q \times f$ value of 129,200 GHz in the series of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics. Above results suggest that the $Q \times f$ value increased by partially replacing Mg^{2+} ions with Zn^{2+} ions. Additionally, a $Q \times f$ value of 33,100 GHz was derived for $\text{Nd}_2\text{Sn}_2\text{O}_7$ ceramics. The observation that the amount of second phase decreased as x increased from 0 to 0.15 implies that the $Q \times f$ value of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ increases as x is increased. However, the $Q \times f$ value of specimens decreased as x increased from 0.10 to 0.15. This was associated with the grain size. Table 1 shows the grain size of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics with various degrees of Zn^{2+} substitution. $\text{Nd}(\text{Mg}_{0.4}\text{Zn}_{0.1}\text{Sn}_{0.5})\text{O}_3$ ceramics had a maximum grain size of 1.59 μm in the series of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics. Moreover, the total number of grain boundaries decreased with an increasing average grain size. According to a previous study, grain boundaries act as plane defects [22]. As a result, the $Q \times f$ of $\text{Nd}(\text{Mg}_{0.4}\text{Zn}_{0.1}\text{Sn}_{0.5})\text{O}_3$

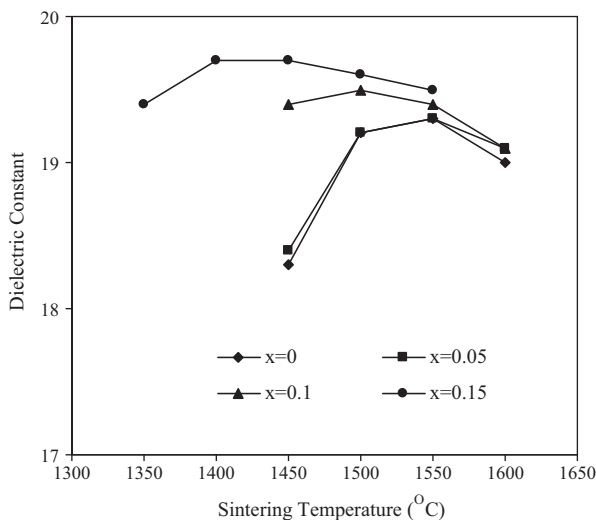


Fig. 4. Dielectric constants of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics with various degrees of Zn^{2+} substitution, following sintering at various temperatures for 4 h.

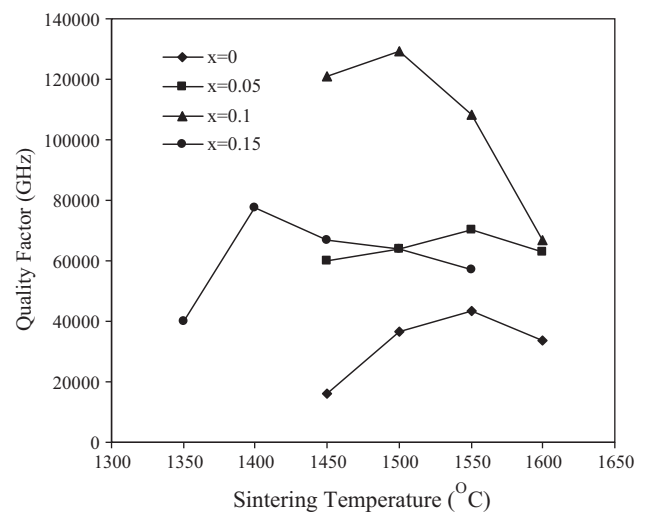


Fig. 5. $Q \times f$ of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics with various degrees of Zn^{2+} substitution, following sintering at various temperatures for 4 h.

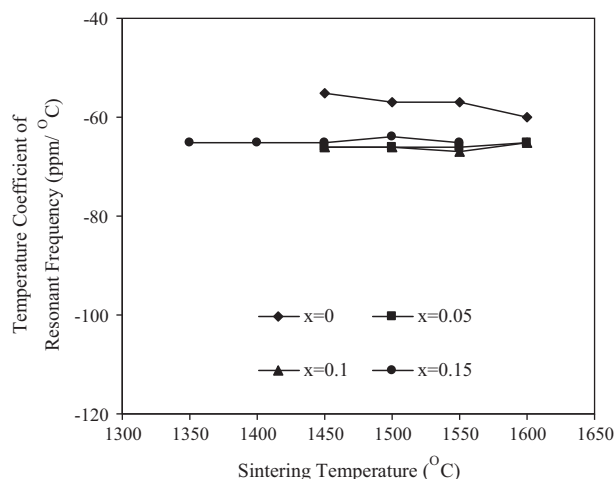


Fig. 6. τ_f of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics with various degrees of Zn^{2+} substitution, following sintering at various temperatures for 4 h.

ceramics obtained a maximum $Q \times f$. Moreover, the $Q \times f$ value of $\text{Nd}(\text{Mg}_{0.35}\text{Zn}_{0.15}\text{Sn}_{0.5})\text{O}_3$ ceramics sintered at 1400 °C was higher than that of $\text{Nd}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics sintered at 1550 °C. Experimental results of $Q \times f$ demonstrated again that substituting of Mg^{2+} with Zn^{2+} can reduce the sintering temperature.

Fig. 6 shows the temperature coefficient of resonant frequency (τ_f) of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics with various degrees of Zn^{2+} substitution, followed by sintering at 1350–1600 °C for 4 h. Generally, τ_f is related to the composition and the degrees of second phase found in ceramics. The observation that the composition of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics with a fixed degree of Zn^{2+} substitution did not vary with sintering temperature implies no significant variation in τ_f of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics with sintering temperature over the entire range of sintering temperatures considered herein. Additionally, the τ_f values of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics were measured as a function of the degrees of Zn^{2+} substitution. Moreover, a τ_f of $-55 \text{ ppm/}^\circ\text{C}$ was obtained for $\text{Nd}_2\text{Sn}_2\text{O}_7$ ceramics. The τ_f value of $\text{Nd}_2\text{Sn}_2\text{O}_7$ ceramics is less negative than that of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics, implying that second phase shifted the τ_f value of the specimen toward a positive direction. Furthermore, the τ_f value of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics became negative as x increased from 0 to 0.05. This observation was associated with a decrease in the amount of second phase as x increased from 0 to 0.05. A τ_f value of $-66 \text{ ppm/}^\circ\text{C}$ was measured for $\text{Nd}(\text{Mg}_{0.4}\text{Zn}_{0.1}\text{Sn}_{0.5})\text{O}_3$ ceramics sintered at 1500 °C for 4 h. A relatively high τ_f value of the $\text{Nd}(\text{Mg}_{0.4}\text{Zn}_{0.1}\text{Sn}_{0.5})\text{O}_3$ ceramics precludes its immediate application potential. Efforts are underway in our laboratory to customize the τ_f value of $\text{Nd}(\text{Mg}_{0.4}\text{Zn}_{0.1}\text{Sn}_{0.5})\text{O}_3$ ceramics.

4. Conclusions

This study elucidated how the degree of Zn^{2+} substitution and sintering temperature affects microwave dielectric properties of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics. The sintering

temperature of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics was reduced by substituting Mg^{2+} with Zn^{2+} . Also, $\text{Nd}(\text{Mg}_{0.4}\text{Zn}_{0.1}\text{Sn}_{0.5})\text{O}_3$ ceramics sintered at 1500 °C for 4 h had a dielectric constant of 19.5, a $Q \times f$ of 129,200 GHz, as well as temperature coefficient of resonant frequency (τ_f) of $-66 \text{ ppm/}^\circ\text{C}$. Experimental results demonstrate that $\text{Nd}(\text{Mg}_{0.4}\text{Zn}_{0.1}\text{Sn}_{0.5})\text{O}_3$ ceramics have a higher dielectric constant and $Q \times f$ than that of $\text{Nd}(\text{Mg}_{0.5}\text{Sn}_{0.5})\text{O}_3$ ceramics. Moreover, the density and amount of second phase affect both the dielectric constant and $Q \times f$ of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics. Furthermore, the dielectric constant and $Q \times f$ of $\text{Nd}(\text{Mg}_{0.5-x}\text{Zn}_x\text{Sn}_{0.5})\text{O}_3$ ceramics were also depended on the ionic polarization and grain size, respectively.

Acknowledgment

The authors would like to thank the National Science Council in Taiwan, for financially supporting this research under Contract No. NSC 100-2622-E-262-001-CC3.

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