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Effect of Sr substitution on microwave dielectric properties of $Nd(Mg_{0.5}Sn_{0.5})O_3$ ceramics

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Abstract

The microwave dielectric properties of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics were examined with a view to their exploitation in mobile communication. The $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics were prepared by the conventional solid-state method with various sintering temperatures. The X-ray diffraction patterns of the $Nd_{2.94/3}Sr_{0.03}(Mg_{0.5}Sn_{0.5})O_3$ ceramics revealed no significant variation of phase with sintering temperatures. A density of 6.88 g/cm^3, a dielectric constant (ε_r) of 19.3, a quality factor ($Q \times f$) of 91,200 GHz, and a temperature coefficient of resonant frequency (τ_f) of -65 ppm/°C were obtained for $Nd_{2.94/3}Sr_{0.03}(Mg_{0.5}Sn_{0.5})O_3$ ceramics that were sintered at 1550 °C for 4 h.

Keywords: $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$; Dielectric constant; Quality factor; Temperature coefficient of resonant frequency

1. Introduction

The benefits of using complex perovskite ceramics are reportedly associated with their excellent microwave dielectric properties [1–3]. 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 properties enable small devices with low loss and high temperature stability to be fabricated. Many investigations of $Nd(Mg_{0.5}Sn_{0.5})O_3$ ceramics have investigated their potential application in resonators, filters, and antennas in modern communication systems, including radars and wireless local area networks, which are operated at microwave frequencies. A dielectric constant of 19.3 and a $Q \times f$ of 43,300 GHz were obtained for $Nd(Mg_{0.5}Sn_{0.5})O_3$ ceramics that were sintered at 1550 °C for 4 h [4]. A dielectric constant of approximately 18.9 and a $Q \times f$ of 32,300 GHz were obtained for 0.25 wt% B_2O_3-doped $Nd(Mg_{0.5}Sn_{0.5})O_3$ ceramics that were sintered at 1500 °C for 4 h [5]. Since the ionic radius of Ba^{2+} (0.135 nm) exceeds that of Ca^{2+} (0.100 nm), $BaLa_4Ti_4O_{15}$ compounds have better microwave dielectric properties than $CaLa_4Ti_4O_{15}$ compounds [6,7]. This fact motivated this study of the effect of the substitution of Nd^{3+} ions (0.0983 nm) by Sr^{2+} ions (0.118 nm) to form $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics.

In this investigation, $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ were synthesized and some of the Nd^{3+} ions were substituted by Sr^{2+} ions to improve their $Q \times f$. The effect of the sintering temperature on the microwave dielectric properties of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics was investigated. $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics were synthesized using the conventional mixed-oxide method and demonstrated to have better microwave dielectric properties than $Nd(Mg_{0.5}Sn_{0.5})O_3$ ceramics. The microwave dielectric properties of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics were found to vary with the extent of Sr^{2+} substitution and sintering temperature. The microwave dielectric properties were further analyzed by densification, X-ray diffraction (XRD) patterns, and observation of their microstructures.

2. Experimental procedure

The starting raw chemicals were highly pure Nd_2O_3 (99.99%), $SrCO_3$ (99.9%), MgO (98.0%), and SnO_2

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(99.0%) powders. The prepared composition was $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ($x=0.02$, 0.03, 0.04). Specimens were prepared using the conventional mixed-oxide method. The starting materials were stoichiometrically weighed. 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 very fine powder thus obtained was then axially pressed at 2000 kg/cm^2 into pellets with a diameter of 11 mm and a thickness of 6 mm. These specimens were then sintered at temperatures of 1450 to 1600 °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 Cu$K\alpha$ radiation (at 30 KV and 20 mA) and a graphite monochromator in the 2θ range of 10°–70°. Scanning electron microscopy (SEM; JEOL JSM-6500 F) and energy dispersive X-ray spectrometry (EDS) were utilized to examine the microstructures of the specimens. The apparent densities of the specimens were measured by Archimedes' method in distilled water. The microwave dielectric properties of the specimens were measured by the postresonator method that was developed by Hakki and Coleman [8]. This method uses a cylindrical specimen of diameter D and length L. The specimens that were used for making microwave dielectric property measurements had an aspect ratio, D/L, of approximately 1.6, which is in the permitted range that was determined by Kobayashi and Katoh [9]. 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. An 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 value of τ_f was measured by the same method as 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/°C) was determined from 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 are the resonant frequencies at T_1 and T_2, respectively.

3. Results and discussion

Fig. 1 shows the X-ray diffraction patterns of $Nd_{2.94/3}Sr_{0.03}(Mg_{0.5}Sn_{0.5})O_3$ ceramics that were sintered at 1450–1600 °C for 4 h. The X-ray diffraction patterns of $Nd_{2.94/3}Sr_{0.03}(Mg_{0.5}Sn_{0.5})O_3$ ceramics did not vary significantly with sintering temperature. Clearly, $Nd_{2.94/3}Sr_{0.03}(Mg_{0.5}Sn_{0.5})O_3$ was the main crystalline phase, which was accompanied by small amount of $Nd_2Sn_2O_7$ as the second phase. $Nd_2Sn_2O_7$ with a cubic crystal structure (ICDD-PDF ♯87-1220) was identified and its formation was difficult prevent in the sample that was prepared by the mixed oxide route. A later investigation will involve preparing of the powder by precipitation from solution. This method may yield a single-phase product. The parameters of interest of a single-phase product potentially have much higher values. As displayed in Fig. 1, the (222) and (400) peaks of the $Nd_2Sn_2O_7$ ceramics were at 29.253° and 33.905°, respectively. All of the peaks of $Nd_{2.94/3}Sr_{0.03}(Mg_{0.5}Sn_{0.5})O_3$ ceramics 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 Miller indices. According to Glazer, the superlattice reflections, with specific combinations of odd (o) and even (e) Miller indices, indicated particular 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), and anti-parallel displacement of A-cations (eeo, eoe, oee) [10]. The extra (1/2(210), 1/2(300), 1/2(320), 1/2 (410), 1/2(421), 1/2(432), and

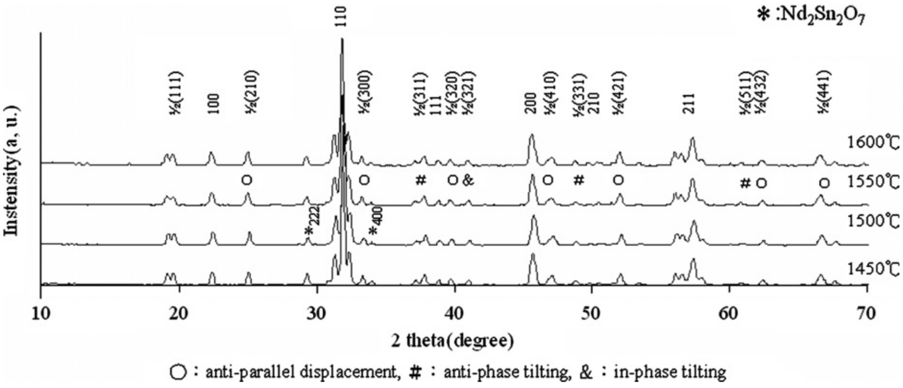

Fig. 1. X-ray diffraction patterns of $Nd_{2.94/3}Sr_{0.03}(Mg_{0.5}Sn_{0.5})O_3$ specimens sintered at 1450–1600 °C for 4 h.

Table 1
Tolerance factor, main phase (%), and second phase (%) of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics.

x	Tolerance factor	Main phase (%)	Second phase (%)
0.02	0.8993	89.05	10.95
0.03	0.9004	93.47	6.53
0.04	0.9015	91.85	8.15

1/2(441)) peaks demonstrate *A*-site cation displacement; the (1/2(311), 1/2(331), and 1/2(511)) peaks indicate anti-phase tilting, and the 1/2(321) peaks indicate in-phase tilting. The 1/2(111) peak is evidence of 1:1 *B*-site cation ordering.

Table 1 shows the tolerance factor and the amounts of main and second phase of the specimens that were sintered at the optimal sintering temperature for 4 h. The tolerance factors of the $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics increased from 0.8993 to 0.9015 as *x* increased from 0.02 to 0.04. The tolerance factors were calculated using the ionic radius data of Shannon [7]. The tolerance factors of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ series were in the anti-phase and in-phase titled region [11], consistent with the X-ray diffraction patterns, that were described above. The amount of the main phase was evaluated from strongest lines of both main and second phases,

$$Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3(\mathrm{vol\%}) = \frac{I_{A(110)}}{I_{A(110)}+I_{B(222)}} \times 100 \quad (2)$$

where I_A and I_B are the strongest lines of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ (110) and $Nd_2Sn_2O_7$ (222), respectively. The amount of the main phase increased from 89.05% to 93.47% as x increased from 0.02 to 0.03, and remaining constant as *x* increased from 0.03 to 0.04. The formation of the second phase of $Nd_2Sn_2O_7$ affected the apparent density and microwave dielectric properties of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics.

Fig. 2 shows the microstructures of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics, following sintering for 4 h at various temperatures. The microstructures of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics when various proportions of Nd^{3+} ions were replaced with Sr^{2+} ions indicated that the average grain size did not vary significantly with the extent of Sr^{2+} ions. The porosities of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics were lower than 1.0% and spanned in the range from 0.5% to 1.0%, following sintering at 1550 °C for 4 h. The $Nd_{2.94/3}Sr_{0.03}(Mg_{0.5}Sn_{0.5})O_3$ 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 $Nd_{2.94/3}Sr_{0.03}(Mg_{0.5}Sn_{0.5})O_3$ ceramics. The pores of $Nd_{2.94/3}Sr_{0.03}(Mg_{0.5}Sn_{0.5})O_3$ ceramics almost disappeared upon sintering at 1550 °C for 4 h. Comparing the microstructures of $Nd_{2.94/3}Sr_{0.03}(Mg_{0.5}Sn_{0.5})O_3$ ceramics that were sintered at different temperatures indicated that the average grain size increased with the sintering temperature. To identify the composition of the second phase, an energy-disperse spectroscopy (EDS) analysis was carried out on the grains of the $Nd_{2.94/3}Sr_{0.03}(Mg_{0.5}Sn_{0.5})O_3$ ceramics that were sintered at 1550 °C for 4 h, as shown in Fig. 2(d). The quantitative analysis, presented in Table 3, reveals that grains A and B were $Nd_{2.94/3}Sr_{0.03}(Mg_{0.5}Sn_{0.5})O_3$ and the grain C was $Nd_2Sn_2O_7$.

$Nd_2Sn_2O_7$ ceramics were synthesized herein using the conventional mixed-oxide method. The effects of the sintering temperature and the sintering time on the microwave dielectric properties of $Nd_2Sn_2O_7$ ceramics were explored. The X-ray diffraction patterns of the $Nd_2Sn_2O_7$ ceramics did not vary significantly with the sintering conditions. The dielectric properties of the $Nd_2Sn_2O_7$ ceramics at microwave frequencies were found to vary with the sintering temperature and sintering time. The $Nd_2Sn_2O_7$ ceramic that was sintered at 1550 °C for 9 h had the highest density, which was 7.11 g/cm^3. Dielectric constants (ε_r) of 16.1–17.0 and quality factors ($Q \times f$) of 30,500–33,100 GHz were obtained following sintering for 9 h at temperatures in the range 1500–1550 °C. Dielectric constants (ε_r) of 16.9–17.0 and quality factors ($Q \times f$) of 28,600–33,100 GHz were obtained upon sintering at 1550 °C for 8–10 h. A dielectric constant (ε_r) of 17.0, a quality factor ($Q \times f$) of 33,100 GHz and a temperature coefficient of resonant frequency (τ_f) of -55 ppm/°C were obtained when $Nd_2Sn_2O_7$ ceramics were sintered at 1550 °C for 9 h, as shown in Table 4.

Fig. 3 shows the apparent densities of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics with different degrees of Sr^{2+} substitution, following sintering at 1450–1600 °C for 4 h. Of the $Nd_{2.94/3}Sr_{0.03}(Mg_{0.5}Sn_{0.5})O_3$ ceramics that were sintered at 1450–1600 °C for 4 h, the one with the highest apparent density was at 1550 °C. $Nd_{2.94/3}Sr_{0.03}(Mg_{0.5}Sn_{0.5})O_3$ ceramic that was sintered at 1550 °C for 4 h had the highest density of 6.88 g/cm^3. The increase in apparent density may have been caused by a decrease in the number of pores, as shown in Fig. 2. The apparent densities of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics that were sintered at 1550 °C for 4 h decreased from 6.89 to 6.87 g/cm^3 as x increased from 0.02 to 0.04. This result was associated with the sum of weights of atoms and the cell volume. Since the weight of an Sr atom is less than that of an Nd atom and the radius of an Sr^{2+} ion is larger than that of an Nd^{3+} ion, the apparent densities of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics are expected to decrease as *x* increase. Table 2 shows the relative densities of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics with different degrees of Sr^{2+} substitution, following sintering at 1550 °C for 4 h. The relative densities of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics decreased from 98.81% to 98.64% as *x* increased from 0.02 to 0.04.

Fig. 3 shows the dielectric constants of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics with different degrees of Sr^{2+} substitution, following sintering at 1450–1600 °C for 4 h. $Nd_{2.94/3}Sr_{0.03}(Mg_{0.5}Sn_{0.5})O_3$ ceramics that were sintered at 1550 °C for 4 h had a dielectric constant of 19.3. The high

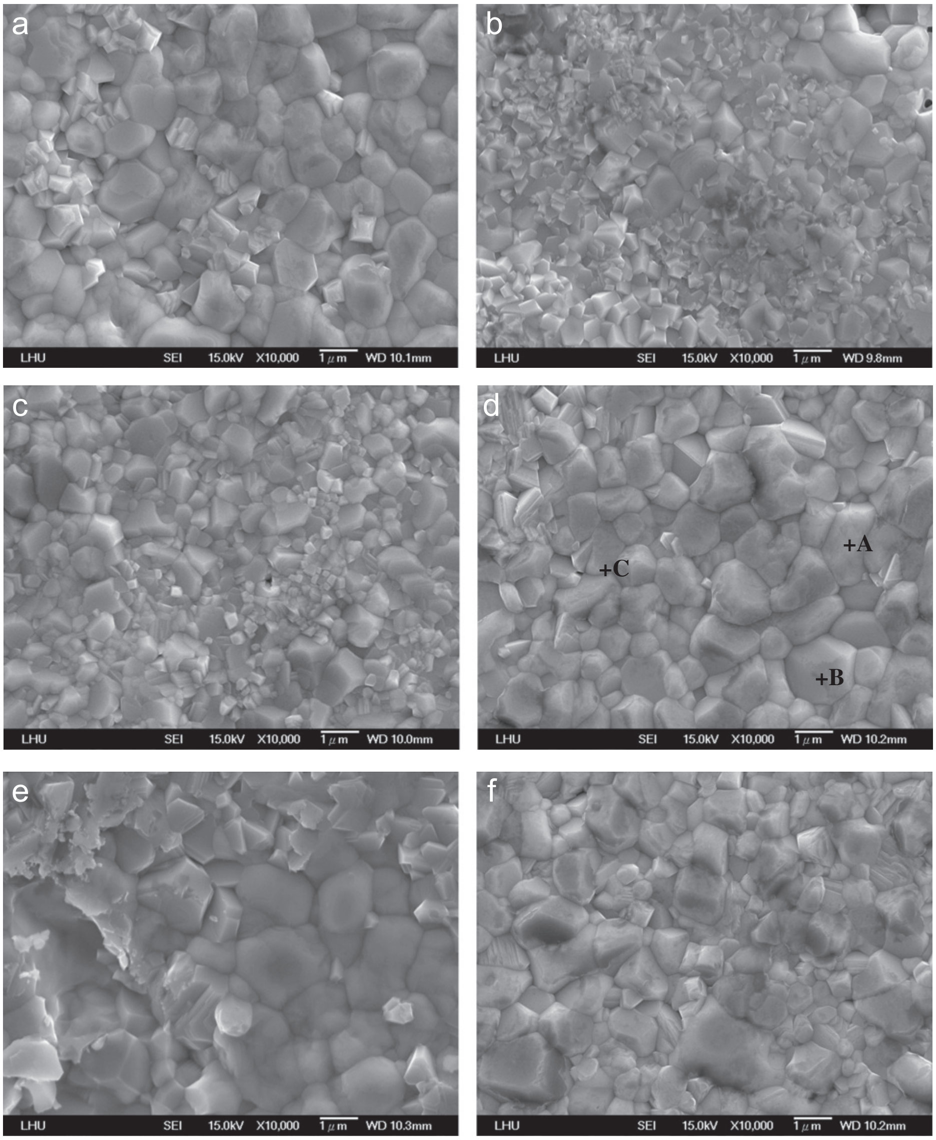

Fig. 2. Microstructures of Nd$_{(1-2x/3)}$Sr$_x$(Mg$_{0.5}$Sn$_{0.5}$)O$_3$ ceramics sintered under different temperatures for 4 h: (a) $x=0.02$/1550 °C, (b) $x=0.03$/1450 °C, (c) $x=0.03$/1500 °C, (d) $x=0.03$/1550 °C, (e) $x=0.03$/1600 °C, (f) $x=0.04$/1550 °C.

Table 2
Relative density, total ionic polarization, and internal strain of Nd$_{(1-2x/3)}$Sr$_x$(Mg$_{0.5}$Sn$_{0.5}$)O$_3$ ceramics.

x	Relative density (%)	Total ionic polarization (A^{o3})	Internal strain
0.02	98.81	13.1286	0.0062
0.03	98.79	13.1354	0.0037
0.04	98.64	13.1422	0.006

Table 3
EDS data of grains of Nd$_{2.94/3}$Sr$_{0.03}$(Mg$_{0.5}$Sn$_{0.5}$)O$_3$ ceramics sintered at 1550 °C for 4 h.

Atomic element	**Nd** (%)	Sr (%)	Mg (%)	Sn (%)	O (%)
A	17.34	0.42	9.06	8.37	64.81
B	14.14	0.94	9.96	7.19	67.77
C	16.25	0	0	14.35	69.40

Table 4
Microwave dielectric properties of $Nd_2Sn_2O_7$ ceramics sintered at 1550 °C for 9 h.

ε_r	$Q \times f$ (GHz)	τ_f (ppm/^{O}C)
17.0	33,100	−55

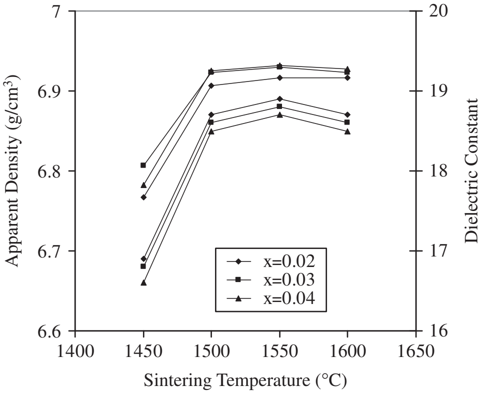

Fig. 3. Apparent densities and dielectric constants of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics with different degrees of Sr^{2+} substitution, following sintering at different temperatures for 4 h.

dielectric constant of $Nd_{2.94/3}Sr_{0.03}(Mg_{0.5}Sn_{0.5})O_3$ ceramics that were sintered at 1550 °C for 4 h did not depend on a high sintering temperature, which decrease in the dielectric constant was associated with drop in the apparent density of the ceramic. A higher density is associated with lower porosity, and, therefore, a higher dielectric constant. The significant decrease in the dielectric constant of $Nd_{2.94/3}Sr_{0.03}(Mg_{0.5}Sn_{0.5})O_3$ ceramics as the sintering temperature decreased from 1500 to 1450 °C was associated with the significant decrease in apparent density and the increase in the number of pores. The dielectric constants of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics that were sintered at 1550 °C for 4 h increased as x increased from 0.02 to 0.04. Many factors, including the second phase and ionic polarization, affect the dielectric constant of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramic. A dielectric constant of 17.0 was obtained for $Nd_2Sn_2O_7$ ceramic, as shown in Table 4. The dielectric constant of the composites can be calculated using the mixture rule,

$$Ln\varepsilon_r = v_1 Ln\varepsilon_{r1} + v_2 Ln\varepsilon_{r2} \tag{3}$$

where ε_r is the dielectric constant of the composite, v_1 and v_2 are the volume fraction of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ and $Nd_2Sn_2O_7$, respectively, and ε_{r1} and ε_{r2} are the dielectric constant of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ and $Nd_2Sn_2O_7$, respectively. The overall dielectric constant is inferred to increase with the amount of the second phase. As shown in Table 1, of the $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics, $Nd_{2.94/3}Sr_{0.03}(Mg_{0.5}Sn_{0.5})O_3$ contained the smallest amount of second phase. Therefore, $Nd_{2.94/3}Sr_{0.03}(Mg_{0.5}Sn_{0.5})O_3$ ceramic is expected to have the highest dielectric constant in the series of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics. However, the dielectric constant of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics increased as x increased from 0.03 to 0.04, owing to ionic polarization. In the case of ionic polarization, as suggested by Tohto *et al.* [6], the dielectric constant can be calculated using the Clausius-Mossotti equation:

$$\varepsilon_r = \frac{3V_m + 8\pi\alpha_D}{3V_m - 4\pi\alpha_D} \tag{4}$$

where Vm represents the molar volume and α_D is the sum of the ionic polarizabilities of individual ions. The dielectric constant that is calculated using Eq. (4) is an intrinsic factor to dielectric property. Dielectric constants therefore depend on the molar volume and ionic polarization. As indicated by Eq. (4), a smaller molar volume or a larger ionic polarization is associated with a larger dielectric constant. Ionic polarization affects dielectric constant much more than does molar volume. Table 2 shows the sum of the ionic polarizations of individual ions of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics with different degrees of Sr^{2+} substitution. The sum of ionic polarizations increased from 13.1286 to 13.1422 A^{o3} as x increased from 0.02 to 0.04 [12], so the dielectric constant of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics increased with x.

Fig. 4 shows the $Q \times f$ of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics with different degrees of Sr^{2+} substitution, following sintering at 1450–1600 °C for 4 h. The relationship between the $Q \times f$ and the sintering temperature of $Nd_{2.94/3}Sr_{0.03}(Mg_{0.5}Sn_{0.5})O_3$ ceramics was consistent with that between the apparent density and the sintering temperature. Many factors affect the microwave dielectric loss, which is composed of intrinsic and extrinsic losses. Intrinsic loss is associated with the vibration modes of the lattice. Extrinsic loss is associated with the density, porosity, second phases, impurities, oxygen vacancies, grain size, and lattice defects [13,14]. Since the $Q \times f$ of $Nd_{2.94/3}Sr_{0.03}(Mg_{0.5}Sn_{0.5})O_3$ ceramics was consistent with the variation of the apparent density, the $Q \times f$ of $Nd_{2.94/3}Sr_{0.03}(Mg_{0.5}Sn_{0.5})O_3$ ceramics is suggested to be dominated

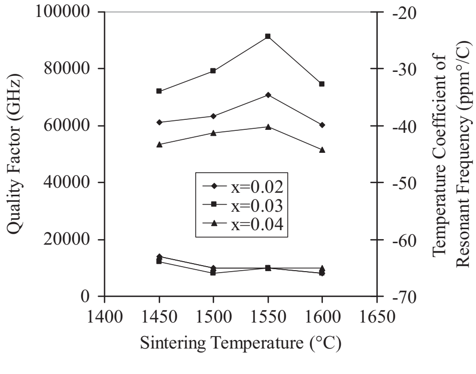

Fig. 4. $Q \times f$ and τ_f of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics with different degrees of Sr^{2+} substitution, following sintering at different temperatures for 4 h.

by the apparent density. In the series of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics, $Nd_{2.94/3}Sr_{0.03}(Mg_{0.5}Sn_{0.5})O_3$ ceramics that were sintered at 1550 °C for 4 h had the highest $Q\times f$ of 91,200 GHz. The results suggest that substituting some Nd^{3+} ions with Sr^{2+} increased $Q\times f$. As indicated in Table 2, the relative densities of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics decreased as x increased. However, in the series of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics, $Nd_{2.94/3}Sr_{0.03}(Mg_{0.5}Sn_{0.5})O_3$ ceramics had the highest $Q\times f$. This result was associated with the second phase and internal strain. $Nd_2Sn_2O_7$ ceramic had a $Q\times f$ of 33,100 GHz, as shown in Table 4. Since $Nd_{2.94/3}Sr_{0.03}(Mg_{0.5}Sn_{0.5})O_3$ ceramic contained the least second phase of any ceramic in the $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ series, it was inferred to have the highest $Q\times f$. The internal strain η can be calculated using following equation, as suggested by Stokes *et al* [15].

$$\beta = 2\eta \tan\theta, \tag{5}$$

where β is the full width at half maximum (FWHM) of an X-ray diffraction peak and θ is the corresponding diffraction angles. The grain size of the specimen also a factor affects FWHM. Since the average grain size did not vary significantly with the extent of substitution of Sr^{2+} ions, Eq. (5) was used for curve fitting to determine the internal strain η of the $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics with different degrees of Sr^{2+} substitution. The curve fitting was based on the least-squares method. In this method, the square of the deviation of the experimental value from the theoretical expectation was calculated while the fitted internal strain was varied. The fitting process was terminated at the fitted internal strain that minimized the deviation. As shown in Table 2, $Nd_{2.94/3}Sr_{0.03}(Mg_{0.5}Sn_{0.5})O_3$ ceramic had the lowest internal strain, 0.037, of any of the $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics. Accordingly, $Nd_{2.94/3}Sr_{0.03}(Mg_{0.5}Sn_{0.5})O_3$ ceramics had the highest $Q\times f$ of the series of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics.

Fig. 4 shows the τ_f of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics with different degrees of Sr^{2+} substitution, following sintering at 1450–1600 °C for 4 h. The negative value of τ_f of the $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics was associated with the in-phase and anti-phase tilting of the octahedral [16]. Generally, τ_f is related to the composition, the amount of additive, and the secondary phases that are present in the ceramics. No significant variation in the τ_f of the $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics with sintering temperature occurred over the entire range of sintering temperature, since the composition of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics with a fixed amount of Sr^{2+} substitution did not vary with sintering temperature. For perovskite ceramics, τ_f is related to tilting of the oxygen octahedron, which is controlled by the tolerance factor of the perovskite ceramics, and, τ_f becomes positive as tolerance factor is increased. However, τ_f did not vary significantly with an increase in the extent of Sr^{2+} substitution in the $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics. This finding is associated with the second phase. As indicated in Table 4, $Nd_2Sn_2O_7$ ceramics had a τ_f of -55 ppm/°C The τ_f of the $Nd_2Sn_2O_7$ ceramics was less negative than that of the $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics, implying that the presence of the second phase shifted the τ_f of the specimen in the positive direction. The $Nd_{2.94/3}Sr_{0.03}(Mg_{0.5}Sn_{0.5})O_3$ ceramic contained the least amount of second phase of any of the $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics. $Nd_{2.94/3}Sr_{0.03}(Mg_{0.5}Sn_{0.5})O_3$ ceramic that was sintered at 1550 °C for 4 h had a τ_f of -65 ppm/°C

4. Conclusions

The effects of the degrees of Sr^{2+} substitution and sintering temperature on the microwave dielectric properties of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics were studied. Substituting Nd^{3+} ions by Sr^{2+} ions was found to improve the $Q\times f$ of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics. $Nd_{2.94/3}Sr_{0.03}(Mg_{0.5}Sn_{0.5})O_3$ ceramics that were sintered at 1550 °C for 4 h had an apparent density of 6.88 g/cm^3, a dielectric constant of 19.3, a $Q\times f$ of 91,200 GHz, and a τ_f of -65 ppm/°C. The formation of $Nd_2Sn_2O_7$ affected the dielectric constant, $Q\times f$, and τ_f of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics. Ionic polarization dominated the dielectric constant of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics. The $Q\times f$ of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics also depended on the density and internal strain. The τ_f of $Nd_{(1-2x/3)}Sr_x(Mg_{0.5}Sn_{0.5})O_3$ ceramics was also affected by the tolerance factor.

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