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Microwave dielectric properties of $Re_3Ga_5O_{12}$ (Re: Nd, Sm, Eu, Dy and Yb) ceramics and effect of TiO_2 on the microwave dielectric properties of $Sm_3Ga_5O_{12}$ ceramics

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Abstract

$Re_3Ga_5O_{12}$ (Re: Nd, Sm, Eu, Dy and Yb) garnet ceramics sintered at 1350–1500 °C had a high quality factor ($Q \times f$) ranging from 40,000 to 192,173 GHz and a low dielectric constant (ε_r) of between 11.5 and 12.5. They also exhibited a relatively stable temperature coefficient of resonant frequency (τ_f) in the range of −33.7 to −12.4 ppm/°C. In order to tailor the τ_f value, TiO_2 was added to the $Sm_3Ga_5O_{12}$ ceramics, which exhibited good microwave dielectric properties. The relative density and grain size increased with addition of TiO_2, resulting in the enhancement of $Q \times f$ value. The τ_f increased with the addition of TiO_2. Excellent microwave dielectric properties of $\varepsilon_r = 12.4$, $Q \times f = 240{,}000$ GHz and $\tau_f = -16.1$ ppm/°C were obtained from the $Sm_3Ga_5O_{12}$ ceramics sintered at 1450 °C for 6 h with 1.0 mol% TiO_2. Therefore, $Re_3Ga_5O_{12}$ ceramics, especially TiO_2-added $Sm_3Ga_5O_{12}$ ceramics are good candidates for advanced substrate materials in microwave integrated circuits (MICs) applications.

Keywords: Sintering; Dielectric properties; Substrates; Garnet structure

1. Introduction

Recently, the development of advanced ceramic substrate materials which are applicable to microwave integrated circuits (MICs) has aroused interest, because of their potential use in the field of microwave telecommunication, in such areas as satellite communications, wireless local area networks and car collision avoidance systems.[1,2] These materials are fundamentally required to have a low dielectric constant (ε_r), in order to minimize the cross-coupling effect with conductors, as well as a high quality factor ($Q \times f$) to increase their frequency selectivity. To obtain the stability of the frequency against temperature changes, a zero temperature coefficient of resonant frequency (τ_f) is also desirable.

Intensive research has been conducted on this subject, and several candidate materials such as alumina (Al_2O_3),[2,3] corundum type ($Mg_4Nb_2O_9$),[4] forsterite (Mg_2SiO_4),[5,6] and willemite (Zn_2SiO_4)[7] ceramics have been suggested. These ceramics have a low ε_r value of less than 12, and a high $Q \times f$ value of more than 200,000 GHz. However, they exhibit a large negative τ_f value. In the case of Al_2O_3, rutile (TiO_2) was added to address this problem, and the resulting $0.9Al_2O_3$–$0.1TiO_2$ ceramics were reported to have good microwave dielectric properties of $\varepsilon_r = 12.4$, $Q \times f = 117{,}000$ GHz and $\tau_f = 1.5$ ppm/°C.[8] In the case of $Mg_4Nb_2O_9$ ceramics with 3.0 wt% LiF, $CaTiO_3$ was additionally added to the system.[9] However, although a zero τ_f value was obtained, the other properties were not desirable. TiO_2 was also added to Mg_2SiO_4 ceramics in order to enhance the τ_f value and lower the sintering temperature. The microwave dielectric properties of the 24.0 wt% TiO_2-added Mg_2SiO_4 sintered at 1200 °C were $\varepsilon_r = 11$, $Q \times f = 82{,}000$ GHz and a near zero τ_f value.[10] With the same purpose, the Zn_2SiO_4–TiO_2 system was investigated, and the result was similar to that obtained for the alumina–titanate system.[7]

Previously, $Nd_3Ga_5O_{12}$ garnet phase was observed as a second phase in the $(1-x)NdGaO_3$–$xCaTiO_3$ system.[11] Although the reported microwave dielectric properties of this system were fairly good and warranted further investigation[11,12], no system-

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atic research on the microwave dielectric properties of garnet materials has yet been performed. Therefore, in this work, rare earth gallium garnet ceramics ($Re_3Ga_5O_{12}$, Re: Nd, Sm, Eu, Dy and Yb) were synthesized and their dielectric properties in the microwave range were studied, in order to evaluate their potential for use as an advanced substrate materials for MIC.

2. Experimental procedure

High purity (>99%) oxide powders were used to synthesize $Re_3Ga_5O_{12}$ (Re: Nd, Sm, Eu, Dy and Yb) ceramics by conventional solid state processing. Nd_2O_3, Sm_2O_3, Eu_2O_3, Dy_2O_3 and Yb_2O_3 (High Purity Chemicals) raw powders were mixed with Ga_2O_3 (High Purity Chemicals, >99.9%) powders stoichiometrically in a nylon jar with zirconia ball for 24 h, and then dried. After drying, calcination was performed at various temperatures for 3 h. After 24–48 h of remilling and subsequent drying, the pellets were pressed hydraulically into a disk-shape and then sintered at 1350–1500 °C for 6 h. To analyze the microstructure of the specimen, X-ray diffraction (XRD: Rigaku D/max-RC, Japan) and scanning electron microscopy (SEM: Hitachi S-4300, Japan) were employed. The relative density was determined by the water immersion method, and the microwave dielectric properties of the specimens were measured by a dielectric resonator technique described by Hakki–Coleman and Courtney.[13,14] The τ_f of the samples was measured in the temperature range from 25 to 90 °C.

3. Results and discussion

Fig. 1(a–c) shows the XRD patterns of the $Re_3Ga_5O_{12}$ (Re: Nd, Sm and Dy) powders calcined at various temperatures. For the $Nd_3Ga_5O_{12}$ specimen, the $Nd_4Ga_2O_9$ phase was formed when Nd_2O_3 and Ga_2O_3 were annealed at 800 °C. As the calcination temperature increased to 1000 °C, an $NdGaO_3$ perovskite phase was found and a homogeneous $Nd_3Ga_5O_{12}$ phase developed without any second phase when the specimens were annealed at 1150 °C. In the case of the $Sm_3Ga_5O_{12}$ specimens, the $Sm_3Ga_5O_{12}$ garnet phase was formed along with a small amount of the $Sm_4Ga_2O_9$ phase for the specimen calcined at 1000 °C. The homogeneous $Sm_3Ga_5O_{12}$ garnet phase developed without any second phase in the specimen calcined at 1150 °C. The perovskite $SmGaO_3$ phase was not found in this specimen. Similar results were also observed for the $Eu_3Ga_5O_{12}$ ceramics. On the other hand, for the $Dy_3Ga_5O_{12}$ specimens, Dy_2O_3 and Ga_2O_3 started to react at 800 °C forming the $Dy_3Ga_5O_{12}$ phase at 1000 °C. The low temperature phases, which were observed in the $Nd_3Ga_5O_{12}$ and $Sm_3Ga_5O_{12}$ specimens, were not observed in the $Dy_3Ga_5O_{12}$ specimen. Similar results were also observed in the $Yb_3Ga_5O_{12}$ specimen. The low temperature phases and their formation temperatures are summarized in Table 1. The above results show that for $Nd_3Ga_5O_{12}$ phase, in which the difference in size between Nd^{3+} and Ga^{3+} ions is large, $Nd_4Ga_2O_9$ and $NdGaO_3$ phases were formed at low temperature, as shown in Fig. 1(a). For the $Dy_3Ga_5O_{12}$ (or $Yb_3Ga_5O_{12}$) phase, in which the difference in size between Dy^{3+} and Ga^{3+} ions is small compared with that of Nd^{3+} and Ga^{3+} ions, no low temperature phase was observed. Therefore, it is considered that the existence of the low temperature phase could be closely related to the difference in size between Re^{3+} and Ga^{3+} ions.

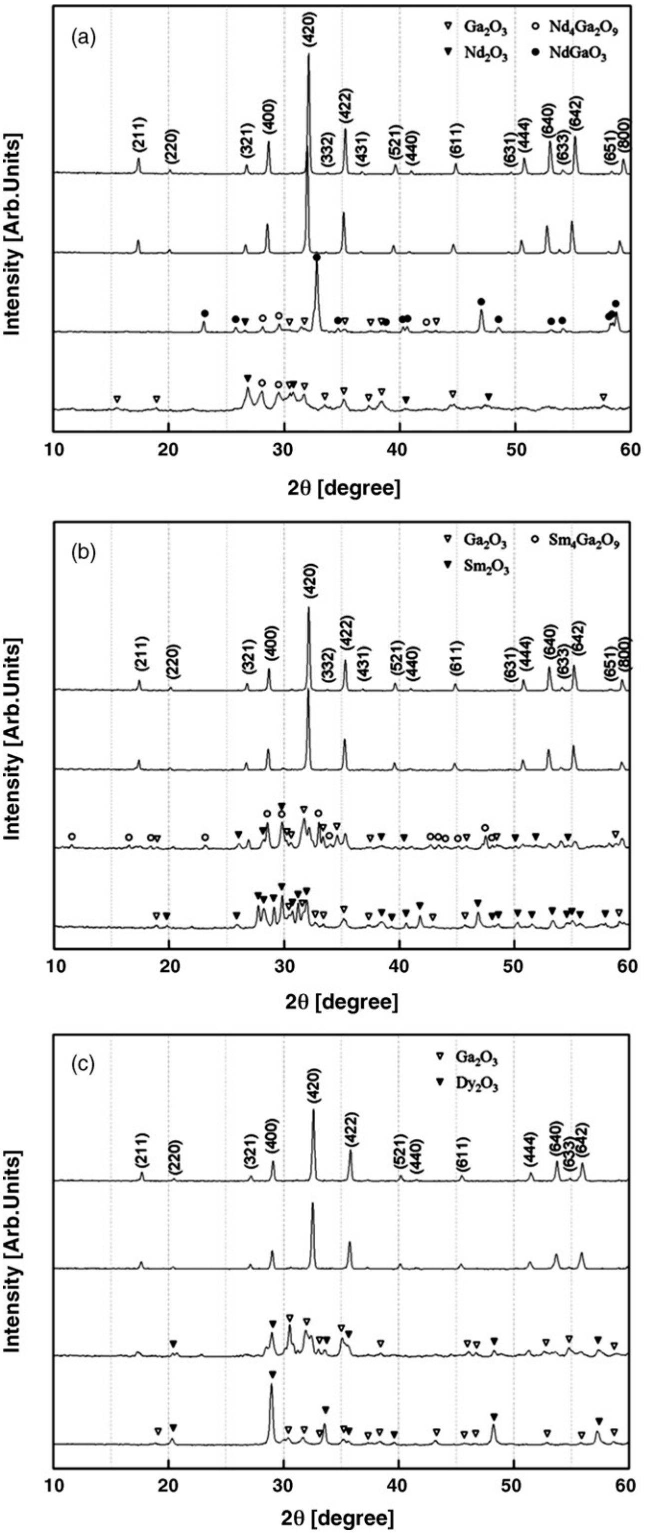

Fig. 1. XRD patterns of the (a) $Nd_3Ga_5O_{12}$, (b) $Sm_3Ga_5O_{12}$ and (c) $Dy_3Ga_5O_{12}$ powders calcined at various temperatures for 3 h.

The relative density and ε_r of the $Re_3Ga_5O_{12}$ (Re: Nd, Sm, Eu, Dy and Yb) ceramics sintered at various temperatures for 6 h

Table 1
The low temperature phases of $Re_3Ga_5O_{12}$ (Re: Nd, Sm, Eu, Dy and Yb) ceramics and their formation temperatures

Re	800 °C	1000 °C	≥1150 °C
Nd	$Nd_4Ga_2O_9$	$NdGaO_3 + Nd_4Ga_2O_9$	$Nd_3Ga_5O_{12}$
Sm	$Sm_2O_3 + Ga_2O_3$	$Sm_3Ga_5O_{12} + Sm_4Ga_2O_9$	$Sm_3Ga_5O_{12}$
Eu	$Eu_2O_3 + Ga_2O_3$	$Eu_3Ga_5O_{12} + Eu_4Ga_2O_9$	$Eu_3Ga_5O_{12}$
Dy	$Dy_2O_3 + Ga_2O_3$	$Dy_3Ga_5O_{12}$	$Dy_3Ga_5O_{12}$
Yb	$Yb_2O_3 + Ga_2O_3$	$Yb_2O_3 + Ga_2O_3$	$Ga_2O_3 + Yb_3Ga_5O_{12}$

are shown in Fig. 2(a and b), respectively. The relative density of the $Nd_3Ga_5O_{12}$ ceramics sintered at 1350 °C was 95.2% of the theoretical density and reached a maximum value of 97.5% of the theoretical density when the specimen was sintered at 1400 °C. For the other specimens sintered at 1350 °C, the relative density was very low, but considerably increased and reached a saturated value of above 95% of the theoretical value for the specimens sintered at 1400 °C. Fig. 2(b) shows the variation of the ε_r value for the $Re_3Ga_5O_{12}$ (Re: Nd, Sm, Eu, Dy and Yb) ceramics sintered at various temperatures. For the $Nd_3Ga_5O_{12}$ ceramics sintered at 1350 °C, the ε_r value was approximately 12.25 and reached a maximum value of 12.4 for the specimen sintered at 1400 °C. The ε_r values of the other garnet ceramics were very low when they were sintered at 1350 °C and significantly increased and reached saturated values ranging from 11.5 to 12.5 when they were sintered at 1400 °C. Therefore, it is considered that the variation of the ε_r value with the sintering temperature was similar to that of the relative density.

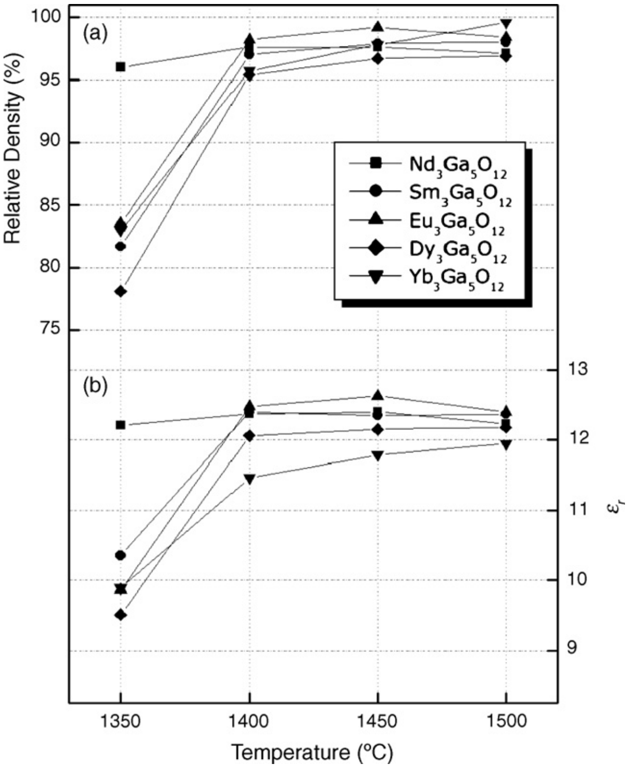

Fig. 2. Variations of: (a) relative density and (b) dielectric constant of the $Re_3Ga_5O_{12}$ (Re: Nd, Sm, Eu, Dy and Yb) ceramics sintered at various temperatures for 6 h.

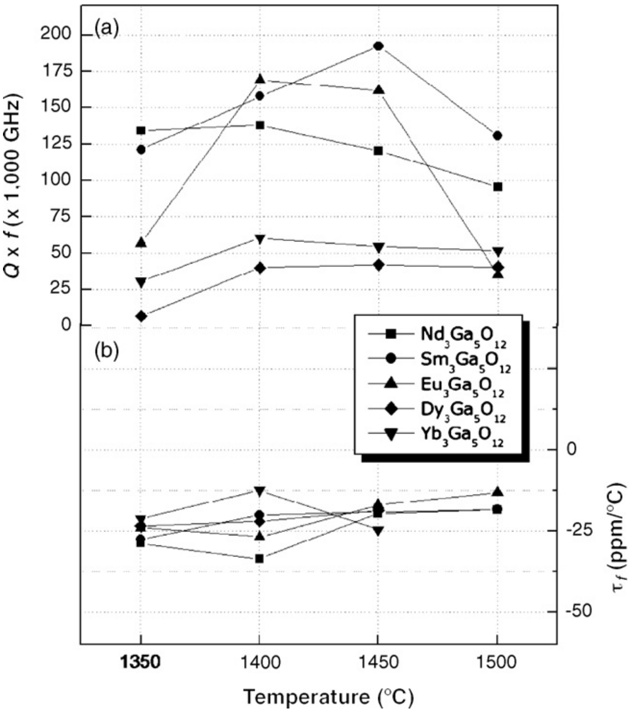

Fig. 3. Variations of: (a) the $Q \times f$ and (b) τ_f values of the $Re_3Ga_5O_{12}$ (Re: Nd, Sm, Eu, Dy and Yb) ceramics sintered at various temperatures for 6 h.

Fig. 3(a) shows the variation of the $Q \times f$ value of the $Re_3Ga_5O_{12}$ (Re: Nd, Sm, Eu, Dy and Yb) ceramics sintered at various temperatures. The $Q \times f$ value of the $Nd_3Ga_5O_{12}$ ceramics sintered at 1350 and 1400 °C was approximately 130,000 GHz, and it decreased when the sintering temperature exceeded 1400 °C. On the other hand, the $Q \times f$ value of the $Sm_3Ga_5O_{12}$ ceramics sintered at 1350 °C was about 120,000 GHz and increased with increasing sintering temperature. A maximum $Q \times f$ value of 192,173 GHz was obtained for the specimen sintered at 1450 °C. For the $Eu_3Ga_5O_{12}$ ceramics sintered at 1350 °C, the $Q \times f$ value was very low, but was significantly increased for the specimen sintered at 1400 °C. The Q-values of the $Dy_3Ga_5O_{12}$ and $Yb_3Ga_5O_{12}$ ceramics are also shown in Fig. 3(a), but they were very low compared with those of $Nd_3Ga_5O_{12}$, $Sm_3Ga_5O_{12}$ and $Eu_3Ga_5O_{12}$ garnet ceramics. The variation of the τ_f value of the $Re_3Ga_5O_{12}$ (Re: Nd, Sm, Eu, Dy and Yb) ceramics sintered at various temperatures is shown in Fig. 3(b). The τ_f values were situated between −35 and −20 ppm/°C and, as such, their variation was not significant. Therefore, the $Nd_3Ga_5O_{12}$, $Sm_3Ga_5O_{12}$ and $Eu_3Ga_5O_{12}$ garnet ceramics have good microwave dielectric properties and, thus, they are promising candidates for advanced substrate materials. In particular, the $Sm_3Ga_5O_{12}$ ceramics sintered at 1450 °C exhibited excellent microwave dielectric properties of $Q \times f = 192{,}173$ GHz, $\varepsilon_r = 12.4$ and $\tau_f = -20$ ppm/°C. However, the τ_f value of the $Sm_3Ga_5O_{12}$ ceramics is relatively large and therefore needs to be reduced.

Since the τ_f value of TiO_2 is approximately 450 ppm/°C, TiO_2 was used to tailor the τ_f value of the $Sm_3Ga_5O_{12}$ ceramics. TiO_2 has often been used to increase the sinterability of ceramics and, therefore, the improvement of the microwave dielectric proper-

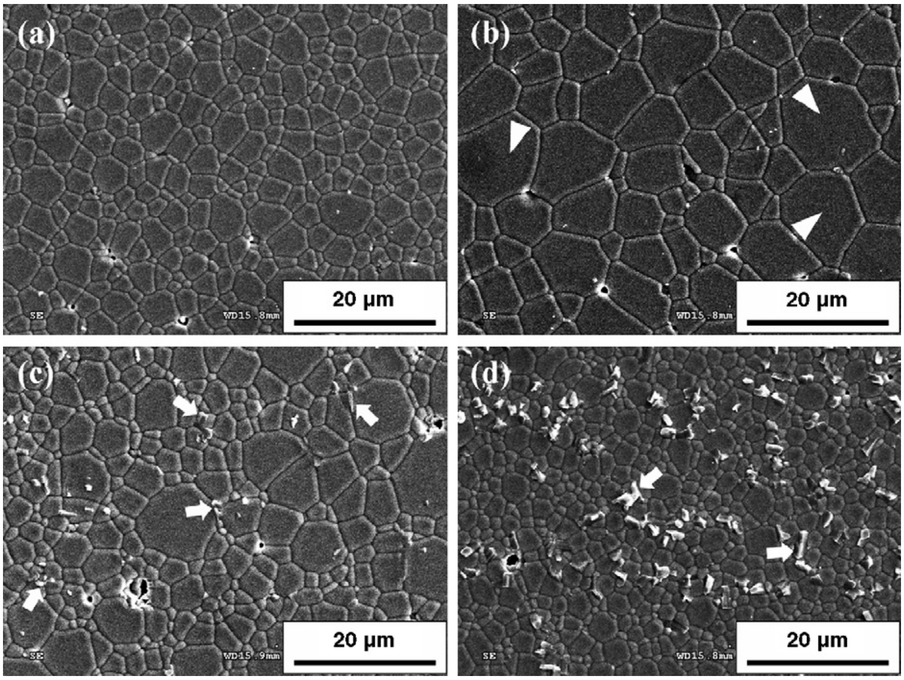

Fig. 4. SEM images of the $Sm_3Ga_5O_{12}$ ceramics containing x mol% of TiO_2 with: (a) $x = 0.0$, (b) $x = 0.02$ and (c) $x = 0.05$ sintered at 1450 °C; and (d) $x = 0.02$ sintered at 1500 °C for 6 h.

ties were also expected through the increase of the sinterability. Fig. 4(a–d) shows the SEM images of the $Sm_3Ga_5O_{12}$ ceramics containing x mol% of TiO_2 with $0.0 \leq x \leq 5.0$. The $Sm_3Ga_5O_{12}$ ceramics sintered at 1450 °C had a dense microstructure without any pores. The average grain size of this specimen was approximately 5.0 µm. Grain growth occurred with the addition of TiO_2 and some of the grain indicated by the arrowhead grew abnormally to a size of 20 µm. A liquid phase was found in the specimen with 5.0 mol% of TiO_2, as indicated by the arrow in Fig. 4(c and d). Therefore, the increase of the grain size could be related to the presence of the liquid phase. Fig. 4(d) shows the SEM image of the $Sm_3Ga_5O_{12}$ ceramics containing 2.0 mol% of TiO_2 sintered at 1500 °C. A large amount of liquid phase was observed in this specimen. It is interesting to note that the grain size of this specimen was smaller than that of the specimen sintered at 1450 °C. According to a previous work, nucleation of the abnormal grain becomes more frequent at higher liquid contents and, thus, a number of the abnormal grains impinge upon each other during the early stages of growth, leading to the development of a microstructure with decreased grain size.[15,16] Therefore, the small grain size for the specimens sintered at 1500 °C could be attributed to the increased amount of liquid phase. EDS analysis was carried out to identify its composition. Fig. 5(a and b) show the EDS spectra taken from the matrix and the liquid phase shown in Fig. 4(d), respectively, and the results of compositional analysis are summarized in Table 2. In the matrix, Sm and Ga ions were detected but the amount of the Ga ion is less than that of nominal composition of $Sm_3Ga_5O_{12}$. On

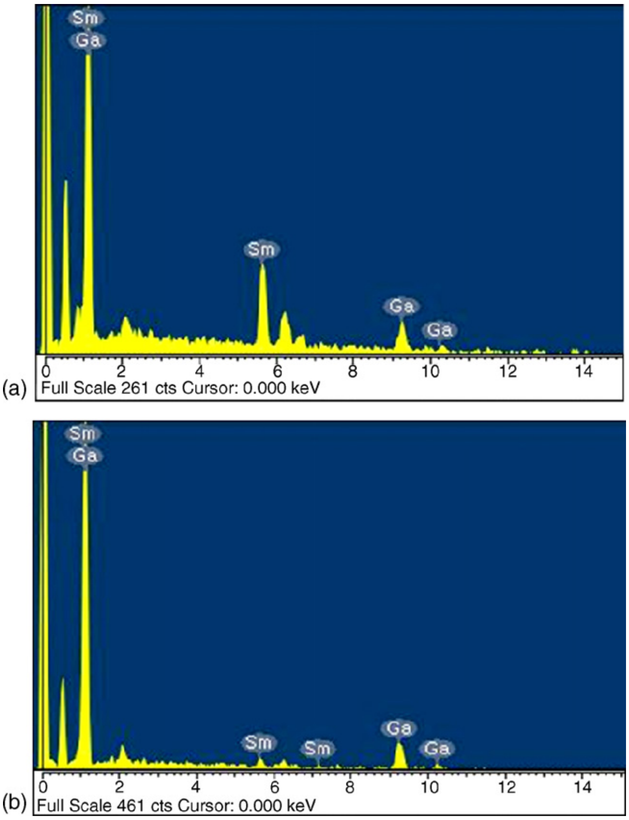

Fig. 5. EDS spectra taken from: (a) the matrix and (b) the liquid phase of the $Sm_3Ga_5O_{12}$ ceramics containing 2.0 mol% of TiO_2 sintered at 1500 °C for 6 h.

Table 2
Chemical compositions of the matrix and the liquid phase of 2.0 mol% TiO_2-added $Sm_3Ga_5O_{12}$ ceramics sintered at 1500 °C for 6 h

Element	Matrix		Liquid phase	
	wt%	at%	wt%	at%
Ga	35.85	54.65	84.18	91.98
Sm	64.15	45.35	15.82	8.02

Table 3
Microwave dielectric properties, resonant frequency, relative density and sintering temperatures of $Re_3Ga_5O_{12}$ (Re: Nd, Sm, Eu, Dy and Yb) and TiO_2-added $Sm_3Ga_5O_{12}$ ceramics

$Re_3Ga_5O_{12}$	Sintering temperature (°C)	Relative density (%)	ε_r	$Q \times f$ (GHz)	τ_f (ppm/°C)	Resonant frequency (GHz)
$Yb_3Ga_5O_{12}$	1400	95.7	11.46	60,294	−12.4	14.9299
$Dy_3Ga_5O_{12}$	1450	96.7	12.15	42,110	−22	14.6103
$Eu_3Ga_5O_{12}$	1400	98.2	12.48	169,155	−17	14.2256
$Sm_3Ga_5O_{12}$	1450	97.9	12.35	192,173	−19.2	13.9448
$Nd_3Ga_5O_{12}$	1400	97.6	12.37	137,811	−33.7	13.7557
TiO_2-added $Sm_3Ga_5O_{12}$	1450	98.6	12.33	234,729	−16.1	14.1459

the other hand, a high concentration of Ga ions was detected in the liquid phase. Therefore, it is considered that some of the Ga ions decomposed from the matrix were used to form the Ga-rich liquid phase.

The variations of the relative density and ε_r, $Q \times f$ and τ_f of the $Sm_3Ga_5O_{12} + x TiO_2$ ceramics with $0.0 \leq x \leq 5.0$ mol% sintered at various temperatures are shown in Fig. 6. The relative densities of all the specimens increased slightly with the addition of a small amount of TiO_2 and decreased when x exceeded 2.0 mol%. However, their variations were not significant and all the specimens had high relative density of more than 95% of the theoretical density. The ε_r value of the specimens increased slightly with the addition of TiO_2 to give a value ranging from 12 to 12.4, but this enhancement was negligible. The variation of the $Q \times f$ value is also shown in Fig. 6. For the specimens sintered above 1450 °C, the $Q \times f$ values decreased with the addition of TiO_2, and this might be due to the increase in the amount of the liquid phase. On the other hand, for the specimens sintered at 1450 °C, the $Q \times f$ value considerably increased with the addition of TiO_2 and showed a maximum value of 240,000 GHz when $x = 1.0$. The enhancement of the $Q \times f$ value could be due to the increase of the relative density and grain size. The $Q \times f$ value considerably decreased with the further addition of TiO_2 and this decrease might be explained by the increase in the amount of the liquid phase. The τ_f value of the $Sm_3Ga_5O_{12}$ ceramics increased with the addition of TiO_2. The $Sm_3Ga_5O_{12}$ ceramics sintered at 1450 °C with 1.0–2.0 mol% of TiO_2, which exhibit a very high $Q \times f$ value, have τ_f values of −16 to −15 ppm/°C. In addition, microwave dielectric properties, resonant frequency, relative density and sintering temperatures of $Re_3Ga_5O_{12}$ (Re: Nd, Sm, Eu, Dy and Yb) and TiO_2-added $Sm_3Ga_5O_{12}$ ceramics are summarized in Table 3. Therefore, $Re_3Ga_5O_{12}$ ceramics, especially TiO_2-added $Sm_3Ga_5O_{12}$ ceramics are good candidates for advanced substrate materials in MIC applications. However, their sintering temperature needs to be decreased for co-firing with metal electrode such Ag and Cu to enable the miniaturization of the microwave devices.

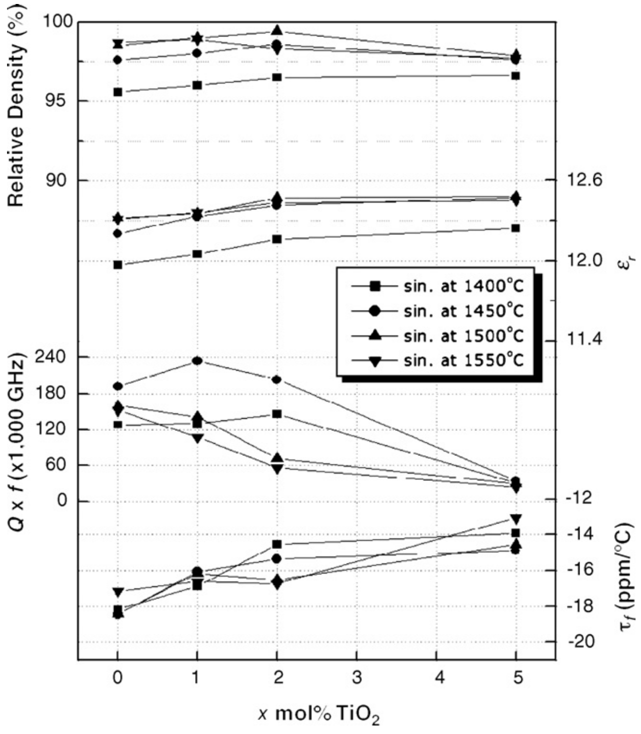

Fig. 6. Variations of the relative density and ε_r, $Q \times f$ and τ_f of the $Sm_3Ga_5O_{12} + x TiO_2$ ceramics with $0.0 \leq x \leq 5.0$ mol% sintered at various temperatures for 6 h.

4. Conclusions

The microwave dielectric properties of $Re_3Ga_5O_{12}$ (Re: Nd, Sm, Eu, Dy and Yb) garnet ceramics were investigated in order to evaluate their potential for use as advanced substrate materials in MICs. A homogeneous $Re_3Ga_5O_{12}$ phase was formed when the specimens were calcined at 1150 °C. All of the specimens had a high relative density (>95% of theoretical density) when they were sintered above 1400 °C. The $Re_3Ga_5O_{12}$ ceramics sintered at 1350–1500 °C exhibited microwave dielectric properties of $40{,}000 \leq Q \times f \leq 192{,}173$ GHz, $11.5 \leq \varepsilon_r \leq 12.5$ and $-33.7 \leq \tau_f \leq -12.4$ ppm/°C. Increases of the grain size and relative density were observed for the $Sm_3Ga_5O_{12}$ ceramics to which a small amount of TiO_2 was added and which were sintered at 1450 °C. These improvements were explained by the presence of the liquid phase, which contains high concentration of Ga_2O_3. The τ_f value was also improved by the addition of TiO_2. The $Sm_3Ga_5O_{12}$ ceramics sintered at 1450 °C for 6 h with 1.0 mol% TiO_2 exhibited improved microwave dielectric properties of $\varepsilon_r = 12.4$, $Q \times f = 240{,}000$ GHz and $\tau_f = -16.1$ ppm/°C.

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