Low temperature sintering and microwave dielectric properties of $Ba_3Ti_5Nb_6O_{28}$ with B_2O_3 and CuO additions

Jeong-Ryeol Kim · Dong-Wan Kim · Sung Hun Yoon · Kug Sun Hong

Abstract The low sintering temperature and the good dielectric properties such as high dielectric constant (ε_r), high quality factor ($Q \times f$), and small temperature coefficient of resonant frequency (TCF) are required for the application of chip passive components in wireless communication low temperature co-fired ceramics (LTCC). In the present study, the sintering behaviors and dielectric properties of $Ba_3Ti_5Nb_6O_{28}$ ceramics were investigated as a function of B_2O_3-CuO content. The pure $Ba_3Ti_5Nb_6O_{28}$ system showed a high sintering temperature (1250°C) and had the good microwave dielectric properties: $Q \times f$ of 10,600 GHz, ε_r of 37, TCF of −12 ppm/°C. The addition of B_2O_3-CuO was revealed to lower the sintering temperature of $Ba_3Ti_5Nb_6O_{28}$, 900°C and to enhance the microwave dielectric properties: $Q \times f$ of 32,500 GHz, ε_r of 40, TCF of 9 ppm/°C. From the X-ray photoelectron spectroscopy (XPS) and X-ray powder diffraction (XRD) studies, these phenomena were explained in terms of the reduction of oxygen vacancies and the formation of secondary phases having the good microwave dielectric properties.

Keywords Low temperature sintering · $Ba_3Ti_5Nb_6O_{28}$ · XPS · Dielectric properties

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1 Introduction

The development of low temperature co-fired ceramics (LTCC) for microwave applications has received much attention, because of the design and functional benefits upon the miniaturization of multilayer devices with high electrical performance by using highly conductive internal electrode metals, such as a silver electrode, with a melting point of 961°C.

There have been three approaches to reduce the sintering temperature of the dielectric ceramics: addition of the glass or the oxides of a low melting-temperature, chemical processing, and utilization of ultra-fine particles for raw materials. In general, liquid phase sintering by adding glass or the oxides of a low melting-temperature such as Li_2CO_3, CuO, PbO, B_2O_3, V_2O_5 and Bi_2O_3 has been known to be effective and the inexpensive way to lower the firing temperature of the ceramics [1–3]. Liquid-phase sintering by adding the oxides of a low melting-temperature is the most inexpensive process among the described method. Yamamoto et al. have suggested that the addition of B_2O_3 or V_2O_5 allows the formation of a large amount of liquid phase [1]. Cho et al. have proposed that $MgTiO_3$-$SrTiO_3$ ceramics with B_2O_3 or CuO have a high quality factor ($Q \times f$) of 75,300 GHz (at 9 GHz), dielectric constant (ε_r) of 19 and temperature coefficient of resonant frequency (TCF) of −8.9 ppm/°C [4]. It also is reported that CuO leads to a lower sintering temperature in TiO_2, $ZnNb_2O_6$, etc [5].

The dielectric properties of $Ba_3Ti_5Nb_6O_{28}$ ceramics have been investigated by Roberts et al. [6]. More recently, Sebastian has investigated the microwave dielectric properties of BaO-TiO_2-Nb_2O_5/Ta_2O_5 system. He has reported that $Ba_3Ti_5Nb_6O_{28}$ ceramic has high $Q \times f$ of 4,500 GHz (at

5.4 GHz), high ε_r of 41 and small TCF of 8 ppm/°C [7]. The sintering temperature of $Ba_3Ti_5Nb_6O_{28}$ was above 1250°C, which is too high to be applicable to LTCC. Unfortunately, though $Ba_3Ti_5Nb_6O_{28}$ ceramic has the good microwave dielectric properties, the effect on the firing temperature and microwave dielectric properties of $Ba_3Ti_5Nb_6O_{28}$ by the addition of dopants has not been thoroughly studied for LTCC applications.

In the present work, B_2O_3 and CuO were chosen as sintering aids to lower the sintering temperature of $Ba_3Ti_5Nb_6O_{28}$ ceramics. The microwave dielectric properties of $Ba_3Ti_5Nb_6O_{28}$ with B_2O_3 and CuO additions associated with the presence of second phases and the absence of oxygen vacancies also have been investigated.

2 Experimental procedure

The $Ba_3Ti_5Nb_6O_{28}$ powders were synthesized by conventional mixed oxide methods: $BaCO_3$ (99.9% pure, Cerac, Milwaukee, WI), TiO_2 and Nb_2O_5 (99.9% pure, High Purity Chemical Laboratory, Saitama, Japan) were mixed homogeneously and calcined at 1150°C for 2 h. The calcined powders containing a proper amount of B_2O_3 and CuO (99.9% pure, High Purity Chemical Laboratory, Saitama, Japan) were ball-milled for 48 h using ethanol solvent. The milled powders were then dried, granulated, and pressed at 1000 kg/cm^2 to yield several disk-type pellets (8 mm in diameter and 4 mm in thickness). The pellets were sintered at 850–1000°C for 2 h with a heating rate of 5°C/min. The bulk density of the sintered samples was determined by the Archimedes method.

Shrinkage of the specimens during heat treatment was measured using a horizontal loading dilatometer with alumina rams and boats (Model DIL402C, Netzsch Instruments, Germany). Polished and thermally etched surfaces of sintered specimens were examined using field emission scanning electron microscopy (FESEM: Model JSM6330F, Japan Electronic Optics Laboratory, Japan). The composition analysis was performed using energy-dispersive spectroscopy (EDS: Model Oxford Link ISIS 300, Oxford Instruments, Bucks, U.K). The formation of second phases was investigated using X-ray powder diffraction (Model M18XHF, Macscience Instruments, Japan).

The microwave dielectric properties of sintered samples were measured at x-band frequencies (8–12 GHz) using a network analyzer (Model HP8720C, Hewlett-Packard, Palo Alto, CA). X-ray photoelectron spectroscopy (XPS) analysis was performed with a VG ESCALAB spectrometer (Model 220i-XL, VG Scientific, Instruments, U.K.). Peak positions were calibrated by taking the C 1s peak (284.6 eV) as a reference.
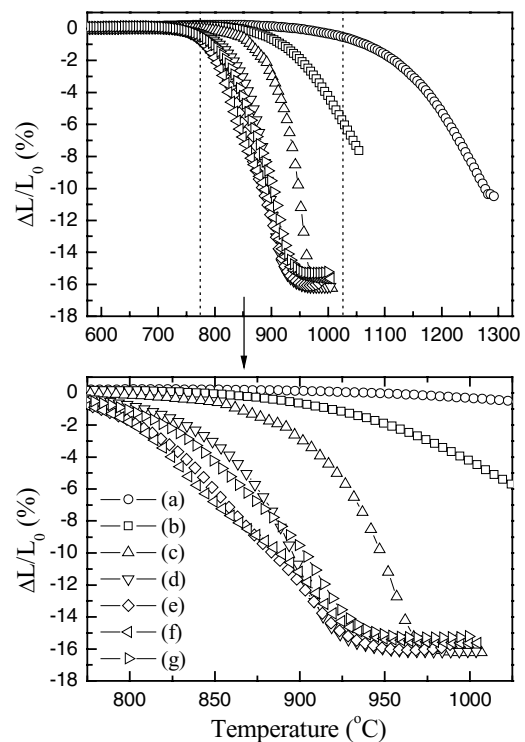

Fig. 1 Shrinkage of $Ba_3Ti_5Nb_6O_{28}$ samples with various contents of B_2O_3 and CuO as a function of temperature: (a) no dopants, (b) 5B, (c) 5C, (d) 3B1C, (e) 2B2C, (f) 1B3C, and (e) 2B5C

3 Results and discussion

3.1 Sintering behavior and phase evolution of low-fired $Ba_3Ti_5Nb_6O_{28}$

Figure 1 shows shrinkage behavior of $Ba_3Ti_5Nb_6O_{28}$ samples with (a) no dopants, (b) 5 wt% B_2O_3 (5B), (c) 5 wt% CuO (5C), (d) 3 wt% B_2O_3 and 1 wt% CuO (3B1C), (e) 2 wt% B_2O_3 and 2 wt% CuO (2B2C), (f) 1 wt% B_2O_3 and 3 wt% CuO (1B3C), and (e) 2 wt% B_2O_3 and 5 wt% CuO (2B5C) as a function of temperature. The shrinkage of $Ba_3Ti_5Nb_6O_{28}$ sample without the dopants appears to occur slowly at ~1000°C and reaches a maximum value at ~1300°C. Though the shrinkage of $Ba_3Ti_5Nb_6O_{28}$ sample with B_2O_3 or CuO occurs at the lower temperature (~900°C) than that of $Ba_3Ti_5Nb_6O_{28}$ sample without the dopants, reaches a maximum value over 1000°C. In contrast, $Ba_3Ti_5Nb_6O_{28}$ samples with both B_2O_3 and CuO exhibit an onset of shrinkage near 750°C, showing the maximum shrinkage at 950°C. In other words, though $Ba_3Ti_5Nb_6O_{28}$ sample with 5B (9.5 vol% B_2O_3) or 5C (3.7 vol% CuO) had the more or the similar amount of sintering aid, the densification of the sample was poorer than $Ba_3Ti_5Nb_6O_{28}$ sample with 1B3C (2.0 vol% B_2O_3 and 2.2 vol% CuO). These results indicate that the co-additions of B_2O_3 and CuO to $Ba_3Ti_5Nb_6O_{28}$ samples can effectively lower the
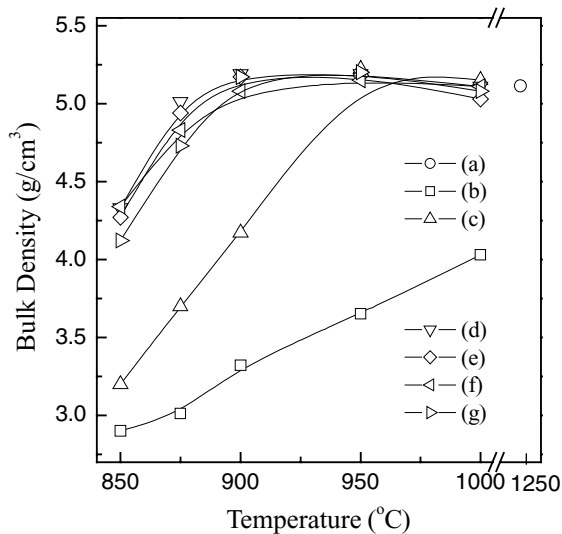
Fig. 2 Change in bulk density of $Ba_3Ti_5Nb_6O_{28}$ samples with various contents of B_2O_3 and CuO as a function of sintering temperature: (a) no dopants, (b) 5B, (c) 5C, (d) 3B1C, (e) 2B2C, (f) 1B3C, and (e) 2B5C

Fig. 3 SEM micrograph of $Ba_3Ti_5Nb_6O_{28}$ samples sintered at 900°C for 2 h with (a) 5B, (b) 5C and (c) 1B3C

sintering temperature of the ceramics. In addition, the higher CuO/B_2O_3 weight ratio as dopants is, the $Ba_3Ti_5Nb_6O_{28}$ samples exhibit an onset of shrinkage at the lower temperature in Fig. 1. On the phase diagram of CuO-B_2O_3 [8], the compositions with a higher B_2O_3 content easily form a liquid phase at the low temperature (450°C), moreover, a eutectic point of $Cu_3B_2O_6$-CuB_2O_4 exists as low as at 880°C. Thus, it can be postulated that the reaction between CuO and B_2O_3 causes the formation of liquid phases and the liquid phases act as the sintering aids for $Ba_3Ti_5Nb_6O_{28}$ samples.

Figure 2 shows the change in bulk density of $Ba_3Ti_5Nb_6O_{28}$ samples with various contents of B_2O_3 and CuO as a function of sintering temperature. The bulk density of $Ba_3Ti_5Nb_6O_{28}$ samples with 5B and 5C sintered at 900°C for 2 h reaches 3.32 and 4.17 g/cm³, respectively. The bulk density of $Ba_3Ti_5Nb_6O_{28}$ samples with 5B and 5C increases with increasing temperature. In contrast, the bulk density of $Ba_3Ti_5Nb_6O_{28}$ samples with both B_2O_3 and CuO sintered at 900°C for 2 h reaches almost 5.15 g/cm³, irrespective of CuO/B_2O_3 ratio. The obtained bulk density of 5.15 g/cm³ corresponds to the bulk density (5.10 g/cm³) of $Ba_3Ti_5Nb_6O_{28}$ sample sintered at 1250°C for 2 h. Therefore, these results reveal that significant reductions in the sintering temperature of $Ba_3Ti_5Nb_6O_{28}$ samples is possible with B_2O_3 and CuO co-additions, while maintaining high density.

The dense microstructure produced by the firing of $Ba_3Ti_5Nb_6O_{28}$ was confirmed by a SEM study which showed a dense microstructure. Figure 3 shows SEM micrographs of $Ba_3Ti_5Nb_6O_{28}$ with various contents of B_2O_3 and CuO sintered at 900°C for 2 h. $Ba_3Ti_5Nb_6O_{28}$ sample with B_2O_3 or CuO shows larger amounts of porosity than $Ba_3Ti_5Nb_6O_{28}$ sample with B_2O_3 and CuO co-addition. It is of interest that low-fired $Ba_3Ti_5Nb_6O_{28}$ sample with both B_2O_3 and CuO
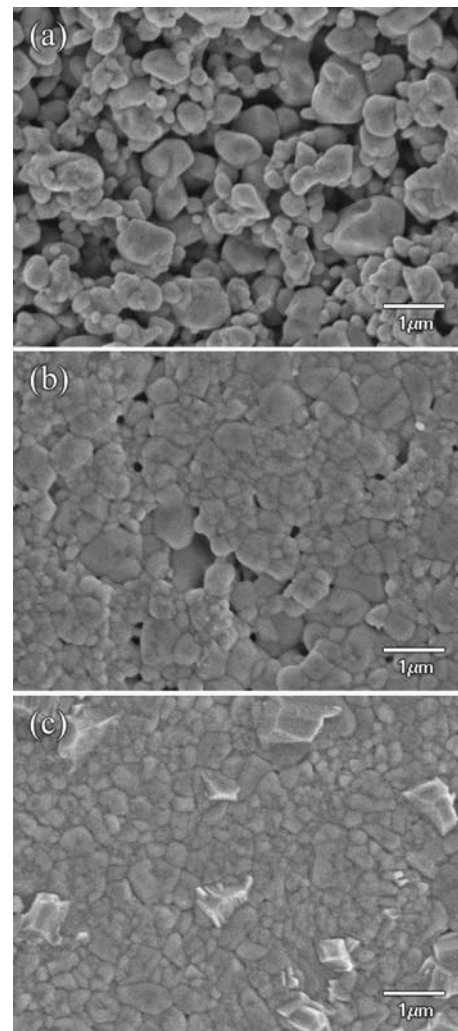
has faceted grains. We found the composition of the faceted grains to be a Cu-rich phase using EDS analysis (not shown here).

Figure 4 shows XRD patterns of (a) $Ba_3Ti_5Nb_6O_{28}$ sample sintered at 1250°C for 2 h and $Ba_3Ti_5Nb_6O_{28}$ samples sintered at 900°C for 2 h with various contents of B_2O_3 and CuO. In Fig. 4, the peaks indicating the presence of a second phase appear in the XRD pattern of the samples containing CuO. The intensity of the second phase peaks slightly increases with increasing amounts of CuO. A small amount of CuO could react with $Ba_3Ti_5Nb_6O_{28}$ and formed $Ba_3Ti_4Nb_4O_{21}$ primarily. Unfortunately, the absence of TiO_2 and Nb_2O_5 remaining after decomposition of $Ba_3Ti_5Nb_6O_{28}$ ceramics is not clear at this moment. XRD pattern of $Ba_3Ti_5Nb_6O_{28}$ sample with 5B indicates no trace of any secondary crystalline phases. We did not found the crystalline phases of B_2O_3-CuO liquid phases, also. These results shows that low-fired $Ba_3Ti_5Nb_6O_{28}$ sample with 1B3C is a mixture of

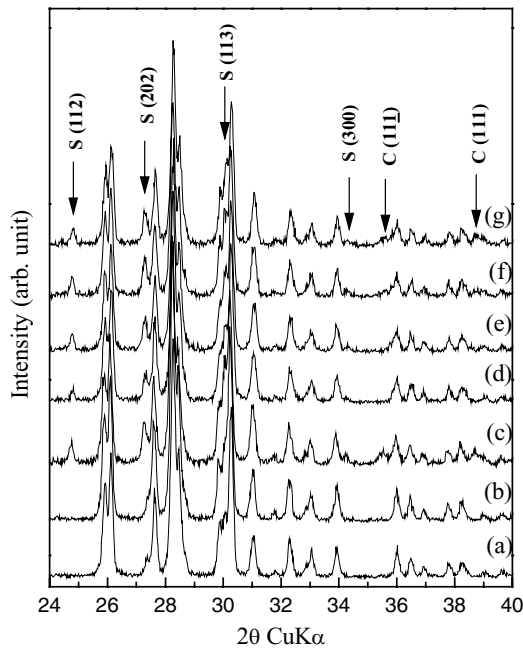**Fig. 4** XRD patterns of (a) $Ba_3Ti_5Nb_6O_{28}$ sample sintered at 1250°C for 2 h and $Ba_3Ti_5Nb_6O_{28}$ samples sintered at 900°C for 2 h with various contents of B_2O_3 and CuO: (b) 5B, (c) 5C, (d) 3B1C, (e) 2B2C, (f) 1B3C, and (e) 2B5C; (S: $Ba_3Ti_4Nb_4O_{21}$, C: CuO)

$Ba_3Ti_5Nb_6O_{28}$, $Ba_3Ti_4Nb_4O_{21}$, remaining CuO and a small amount of liquid phases, mainly.

3.2 Microwave dielectric properties of low-fired $Ba_3Ti_5Nb_6O_{28}$

The dielectric properties of $Ba_3Ti_5Nb_6O_{28}$ have been initially reported by Reberts et al. [6], and more recently, Sebastian has reported the microwave properties of BaO-TiO_2-Nb_2O_5/Ta_2O_5 system [7]. The microwave properties of $Ba_3Ti_5Nb_6O_{28}$, $Ba_3Ti_4Nb_4O_{21}$ and low-fired $Ba_3Ti_5Nb_6O_{28}$ are summarized in Table 1. It is postulated that the microwave dielectric properties of low-fired $Ba_3Ti_5Nb_6O_{28}$ samples are affected by the presence of second phases shown in Fig. 4. In Table 1, ε_r of low-fired $Ba_3Ti_5Nb_6O_{28}$ with B_2O_3 or CuO shows decreased value

from that (37.0) of pure $Ba_3Ti_5Nb_6O_{28}$ sintered at 1250°C for 2 h, because of the poor densification at low temperatures. In contrast, ε_r (~ 40) of low-fired $Ba_3Ti_5Nb_6O_{28}$ samples with both B_2O_3 and CuO co-addition is higher than that of low-fired $Ba_3Ti_5Nb_6O_{28}$ with B_2O_3 or CuO, because of high densification. It is noteworthy that ε_r of low-fired $Ba_3Ti_5Nb_6O_{28}$ samples with B_2O_3 and CuO co-addition is higher even than that of pure $Ba_3Ti_5Nb_6O_{28}$ densified fully. In low-fired $Ba_3Ti_5Nb_6O_{28}$ samples, the second phases are $Ba_3Ti_4Nb_4O_{21}$, remaining CuO and a small amount of liquid phases, mainly. $Ba_3Ti_4Nb_4O_{21}$ sintered at 1270°C for 4 h had a high ε_r($\sim$55) [7], and CuO had a very low ε_r($\sim$11) [9]. Although the dielectric properties of liquid phase are not fully characterized, liquid phase has a low ε_r, in general. Because of much more $Ba_3Ti_4Nb_4O_{21}$ than other phases (Fig. 4), $Ba_3Ti_4Nb_4O_{21}$ phase would result in the slight increase of ε_r in low-fired $Ba_3Ti_5Nb_6O_{28}$ with B_2O_3 and CuO co-addition. $Ba_3Ti_4Nb_4O_{21}$ phase having a large TCF (100 ppm/°C) [7] would contribute to the slight increase in TCF of low-fired $Ba_3Ti_5Nb_6O_{28}$ with B_2O_3 and CuO co-addition, also.

Low-fired $Ba_3Ti_5Nb_6O_{28}$ samples with both B_2O_3 and CuO possess a higher $Q \times f$ over 32,000 GHz than expected. In general, the addition of additives for low temperature sintering is accompanied by a significant decrease in the microwave dielectric properties. It is noteworthy that $Q \times f$ of low-fired $Ba_3Ti_5Nb_6O_{28}$ samples is higher than that of pure $Ba_3Ti_5Nb_6O_{28}$ densified fully. Though $Ba_3Ti_4Nb_4O_{21}$ phase has a relatively high $Q \times f$ value (9,500 GHz) [7], the $Q \times f$ value is not as high as it enhances the $Q \times f$ of low-fired $Ba_3Ti_5Nb_6O_{28}$ samples over 32,000 GHz. Dielectric losses fall into two categories, intrinsic and extrinsic [10]. In general, extrinsic losses are associated with imperfections on the crystal structure, such as defects, grain boundaries, porosity, micro-cracks, etc. Because the low-fired $Ba_3Ti_5Nb_6O_{28}$ samples with B_2O_3 and CuO have the similar microstructure and density to the pure $Ba_3Ti_5Nb_6O_{28}$ sample, the diminishment in defects such as oxygen vacancy could largely enhance the quality factor. Deterioration in the quality factor that is associated with defects in titanium-containing ceramics (including reduction) has been reported in various publications [11–13].

Table 1 Microwave dielectric properties of $Ba_3Ti_5Nb_6O_{28}$, $Ba_3Ti_4Nb_4O_{21}$ and low-fired $Ba_3Ti_5Nb_6O_{28}$ ceramics

Composition	Sintering conditions	ε_r	$Q \times f$ (GHz)	TCF (ppm/°C)
$Ba_3Ti_5Nb_6O_{28}^*$	1300°C for 4 h	41	4,500	8
$Ba_3Ti_5Nb_6O_{28}$	1250°C for 2 h	37.0	10,600	−12
$Ba_3Ti_4Nb_4O_{21}^*$	1270°C for 4 h	55	9,500	100
$Ba_3Ti_5Nb_6O_{28}$ with 5B	900°C for 2 h	15.9	14,000	−13
$Ba_3Ti_5Nb_6O_{28}$ with 5C	900°C for 2 h	26.6	14,100	21
$Ba_3Ti_5Nb_6O_{28}$ with 3B1C	900°C for 2 h	38.6	29,800	5
$Ba_3Ti_5Nb_6O_{28}$ with 2B2C	900°C for 2 h	40.2	32,200	5
$Ba_3Ti_5Nb_6O_{28}$ with 1B3C	900°C for 2 h	40.3	32,500	9
$Ba_3Ti_5Nb_6O_{28}$ with 2B5C	900°C for 2 h	39.0	32,300	11

* Reference 7.
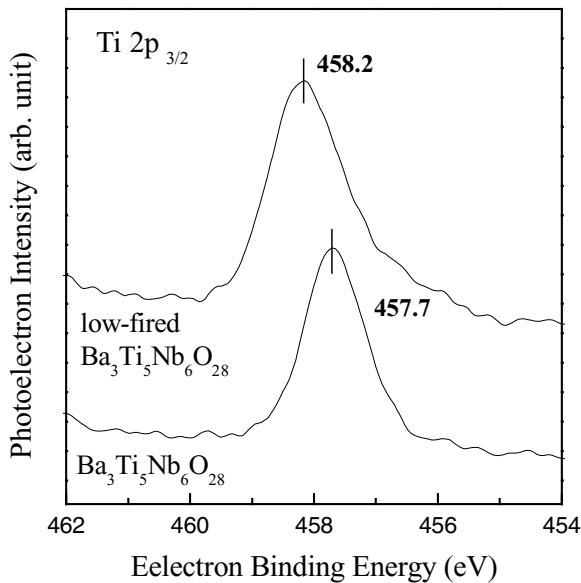
Fig. 5 XPS spectra of Ti 2p$_{2/3}$ for Ba$_3$Ti$_5$Nb$_6$O$_{28}$ sample sintered at 1250°C for 2 h in air and Ba$_3$Ti$_5$Nb$_6$O$_{28}$ sample with 1B3C sintered at 900°C in air

Figure 5 shows Ti 2p$_{3/2}$ spectra of Ba$_3$Ti$_5$Nb$_6$O$_{28}$ sample sintered at 1250°C and Ba$_3$Ti$_5$Nb$_6$O$_{28}$ sample with 1B3C sintered at 900°C in air. The binding energy of Ti 2p$_{3/2}$ peak for Ba$_3$Ti$_5$Nb$_6$O$_{28}$ sample sintered at 1250°C and Ba$_3$Ti$_5$Nb$_6$O$_{28}$ sample with 1B3C sintered at 900°C in air is 457.7 and 458.2 eV, respectively. The binding energy of Ti 2p$_{3/2}$ peak for Ti^{3+} is lower than that for Ti^{4+}, due to binding with less oxygens. For example, Moulder has reported that the binding energy of Ti 2p$_{3/2}$ peak for Ti$_2$O$_3$ and TiO$_2$ is 457.3 and 458.8 eV, respectively [14]. These results can indicate that the defects such as oxygen vacancies (reaction (1)) decrease in low-fired Ba$_3$Ti$_5$Nb$_6$O$_{28}$ samples.

$$2\text{Ti}_{Ti}^{X} + O_O^{X} \Leftrightarrow 2\text{Ti}'_{Ti} + V_O^{\bullet\bullet} + \frac{1}{2}O_2 \qquad (1)$$
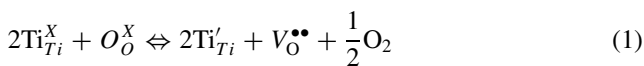
The reduction of defects in low-fired Ba$_3$Ti$_5$Nb$_6$O$_{28}$ samples could result from the low sintering temperature and ionic compensation. While Ti^{4+} can be slightly oxygen-deficient and the diffusion coefficient can be too low to allow oxygenation of thick samples at high temperature [14], the phenomena do not occur at low temperature ($\sim$ 900°C). Also, Templeton et al. [14] reported that the addition of divalent and trivalent dopant (i.e. Cu^{2+}) ions with an ionic radius similar to Ti^{4+} ion, resulted in a TiO$_2$ ceramic with an increased quality factor. Similarly, in low-fired Ba$_3$Ti$_5$Nb$_6$O$_{28}$ samples, reduction of the Ti^{4+} ion can be prevented by a favorable compensation mechanism when CuO is added. Therefore, the increase in the $Q \times f$ of Ba$_3$Ti$_5$Nb$_6$O$_{28}$ samples fully densified at low temperature can be attributed to the absence of oxygen vacancies, i.e., Ti^{4+} reductions.

4 Conclusion

The sintering behaviors and dielectric properties of Ba$_3$Ti$_5$Nb$_6$O$_{28}$ ceramics were investigated as a function of B$_2$O$_3$-CuO content. It was found that simultaneous additions of both B$_2$O$_3$ and CuO to Ba$_3$Ti$_5$Nb$_6$O$_{28}$ enabled a reduction in sintering temperature from 1250°C to 900°C. This can be attributed to the formation of B$_2$O$_3$-CuO liquid phases. During sintering of Ba$_3$Ti$_5$Nb$_6$O$_{28}$ samples with both B$_2$O$_3$ and CuO, several second phases were observed. The major second phase is Ba$_3$Ti$_4$Nb$_4$O$_{21}$. Ba$_3$Ti$_4$Nb$_4$O$_{21}$ having high ε_r (55) and TCF (100 ppm/°C) enhanced the ε_r and TCF of low-fired Ba$_3$Ti$_5$Nb$_6$O$_{28}$ ceramics. The drastic increase in the $Q \times f$ of low-fired Ba$_3$Ti$_5$Nb$_6$O$_{28}$ samples with B$_2$O$_3$ and CuO co-addition is interpreted to be due to rather good properties of second phase, high densification and less oxygen vacancies than in Ba$_3$Ti$_5$Nb$_6$O$_{28}$ samples sintered at 1250°C. Therefore, Ba$_3$Ti$_5$Nb$_6$O$_{28}$ with both B$_2$O$_3$ and CuO can be a suitable candidate for low temperature co-fired ceramic (LTCC), due to its reduced sintering temperature and good properties: $Q \times f = 32, 500$ GHz, $\varepsilon_r = 40$, TCF = 9 ppm/°C.

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