



# Dielectric Properties and Sintering Characteristics of $\text{CaTiO}_3\text{-(Li}_{1/2}\text{Nd}_{1/2})\text{TiO}_3$ Ceramics

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**Abstract.** The dielectric properties and the sintering effect upon microstructure of  $(1-x)\text{CaTiO}_3\text{-}x(\text{Li}_{1/2}\text{Nd}_{1/2})\text{-TiO}_3$  Ceramics are investigated in this paper.  $\text{Nd}^{3+}$  and  $\text{Mg}^{2+}$  ions co-substitution for  $\text{Ca}^{2+}$  on A site improves the sintering characteristic of  $\text{CaTiO}_3$  ceramics with forming orthorhombic perovskite structure. The structure of  $(1-x)\text{CaTiO}_3\text{-}x(\text{Li}_{1/2}\text{Nd}_{1/2})\text{TiO}_3$  changes from orthorhombic to tetragonal as  $(\text{Li}_{1/2}\text{Nd}_{1/2})\text{TiO}_3$  addition increasing. Limited solubility of  $(\text{Li}_{1/2}\text{Nd}_{1/2})\text{TiO}_3$  in  $\text{CaTiO}_3$  forming a part solid solution compound achieves the adjustment of  $\tau_\varepsilon$  for  $\text{CaTiO}_3$  at low sintering temperature. The proper dielectric properties with  $\varepsilon = 78$ ,  $\tan \delta = 0.0006$ ,  $\tau_\varepsilon = +7 \text{ ppm}/^\circ\text{C}$  are obtained for  $0.8\text{Ca}_{0.67}(\text{Nd,Mg})_{0.22}\text{TiO}_3\text{-}0.2(\text{Li}_{1/2}\text{Nd}_{1/2})\text{TiO}_3$  ceramics.

**Keywords:** sintering, perovskite structure,  $\text{CaTiO}_3$ , dielectric performances

## 1. Introduction

Microwave communication has been rapid development in recent years, the key components, such as filters and oscillators must have high dielectric constant, low dielectric loss and near zero temperature coefficient of resonance frequency. Hitherto, several materials with high dielectric constant have been reported and most of them are focused on plural elements system including  $\text{TiO}_2$ , such as  $\text{BaO-Nd}_2\text{O}_3\text{-TiO}_2\text{-Ta}_2\text{O}_5$  [1, 2],  $\text{BaO-Sm}_2\text{O}_3\text{-TiO}_2$  [3, 4],  $\text{CaO-Li}_2\text{O-Ln}_2\text{O}_3\text{-TiO}_2$  [5–9], or  $(\text{A}_{1/2}^{+1}\text{A}_{1/2}^{+3})\text{TiO}_3$  system [10]. Among of these materials, compositions with perovskite structure based on  $(\text{A}_{1/2}^{+1}\text{A}_{1/2}^{+3})\text{TiO}_3$  system or  $\text{CaTiO}_3$  system have superior microwave properties:  $\varepsilon = 81$ ,  $Qf = 6150 \text{ GHz}$ ,  $\tau_f = +17 \text{ ppm}/^\circ\text{C}$  for  $0.6(\text{Li}_{1/2}\text{Sm}_{1/2})\text{TiO}_3\text{-}0.4(\text{Na}_{1/2}\text{Sm}_{1/2})\text{TiO}_3$  ceramics [10],  $\varepsilon = 110$ ,  $Qf = 4500 \text{ GHz}$ ,  $\tau_f = +7 \text{ ppm}/^\circ\text{C}$  for  $\text{CaO-SrO-Li}_2\text{O-Ln}_2\text{O}_3\text{-TiO}_2$  ( $\text{Ln} = \text{Sm, Nd}$ ) system [5],  $\varepsilon 108, Qf = 17200 \text{ GHz}$  in  $(\text{Ca}_{1-x}\text{Nd}_{2x/3})\text{TiO}_3$  system [11] and  $0.67\text{Ca}_{2/5}\text{Sm}_{2/5}\text{TiO}_3\text{-}0.33\text{Li}_{1/2}\text{Sm}_{1/2}\text{TiO}_3$  ceramics with  $\varepsilon = 95.5$ ,  $Qf = 7200 \text{ GHz}$ ,  $\tau_f = 0 \text{ ppm}/^\circ\text{C}$  [9].

$\text{CaTiO}_3$  and  $(\text{A}_{1/2}^{+1}\text{A}_{1/2}^{+3})\text{TiO}_3$  are promising materials with high dielectric constant and high  $Qf$  value.

$\text{CaTiO}_3$  is of orthorhombic perovskite structure [11],  $\text{Li}_{1/2}\text{Ln}_{1/2}\text{TiO}_3$  ( $\text{Ln} = \text{Sm, Nd}$ ) is of orthorhombic perovskite structure (or cubic perovskite structure) [10], both have high dielectric constant and show positive and negative temperature coefficient of frequency, respectively, but both of them have the same drawback: poor sintering behavior.

Masashi Yoshida et al. [11] have succeeded in synthesizing  $\text{CaTiO}_3$  ceramics with Nd substitution for Ca and obtained good microwave properties ( $\varepsilon = 108$ ,  $Qf = 17200 \text{ GHz}$ ). They pointed that structure of  $\text{CaTiO}_3$  varied with Nd substitution from  $\text{GdFeO}_3$ -type structure to  $\text{La}_{2/3}\text{TiO}_3$ -type double-layered structure, and indicated the relationship between dielectric properties and structure change, but they did not mention the affection of microstructure change on the temperature characteristic of resonance frequency.

The purpose of present work is to synthesize densification  $\text{CaTiO}_3$  ceramics with perovskite structure by adding Nd ion and Mg ion co-substitution for Ca ion, then combined  $\text{Li}_{1/2}\text{Nd}_{1/2}\text{TiO}_3$  to form solid solution compound of  $\text{CaTiO}_3\text{-Li}_{1/2}\text{Nd}_{1/2}\text{TiO}_3$ , investigating its dielectric properties and sintering characteristics or microstructure in achieving the proper dielectric constant,

low dielectric loss and stable temperature coefficient of dielectric constant.

## 2. Experiment Procedures

CaCO<sub>3</sub>, Li<sub>2</sub>CO<sub>3</sub>, Nd<sub>2</sub>O<sub>3</sub>, MgO and TiO<sub>2</sub> with high purity (>99.5%) were used as starting materials and the dielectric samples were synthesized by the conventional solid-state reaction method. The powders were weighed according to the composition Ca<sub>1-y</sub>M<sub>2y/3</sub>TiO<sub>3</sub> (y = 0.0, 0.1, 0.15, 0.2, 0.25, 0.3, 0.33, 0.35, 0.4, M = Nd + Mg, Nd/Mg = 4:1, atom ratio) and Li<sub>1/2</sub>Nd<sub>1/2</sub>TiO<sub>3</sub>, and milled with ZrO<sub>2</sub> balls for 24 h in ethyl alcohol to prevent dissolution of Li<sub>2</sub>CO<sub>3</sub> in water, then dried and sieved using 120 mesh screen. The Ca<sub>1-y</sub>M<sub>2y/3</sub>TiO<sub>3</sub> powders was calcined at 900°–1260°C for 3 h and Li<sub>1/2</sub>Nd<sub>1/2</sub>TiO<sub>3</sub> was calcined at 825° for 2 h in air, respectively. These calcined powders were weighed again according to the formulas of (1 - x) Ca<sub>1-y</sub>M<sub>2y/3</sub>TiO<sub>3</sub> - xLi<sub>1/2</sub>Nd<sub>1/2</sub>TiO<sub>3</sub>, then re-milled with ZrO<sub>2</sub> balls for 24 h in ethyl alcohol. After dried, the powders with 5 wt% polyvinyl alcohol as a binder were pressed into rods of 13 mm in diameter and 2–3 mm in thickness under a pressure of 15 Mpa. These pellets were heated at 600° for 2 h to eliminate the binder, then sintered at 1180–1280° for 3 h in air.

X-ray powder diffraction with Cu K<sub>α</sub> radiation was used to determine the crystalline phases and Scanning Electron Microscope evaluate the microstructures. The dielectric constant and dielectric loss were measured by using HP4192B LCR meters from 10 KHz to 1 MHz, and the temperature coefficient of dielectric constant was measured using an impedance analyzer equipped with a thermostat in the range from room temperature to 85°. The bulk densities of the sintered pellets were determined by Archimedes method.

## 3. Results and Discussion

As we know, it is very difficult to prepare pure CaTiO<sub>3</sub> phase by conventional solid-state reaction method as there are three compounds in CaO-TiO<sub>2</sub> system: CaTiO<sub>3</sub>, Ca<sub>4</sub>Ti<sub>3</sub>O<sub>10</sub>, and Ca<sub>3</sub>Ti<sub>2</sub>O<sub>7</sub>. The latter both compounds are dissimilarity dissolvent compound with lower melting point than that of CaTiO<sub>3</sub>, which is impeditive to form pure CaTiO<sub>3</sub> phase. In previous reports sintered sample of CaTiO<sub>3</sub> phase could be obtained by unequal valence substituting on A or B site [11, 12].

In our work, Nd<sup>3+</sup> ion and Mg<sup>2+</sup> ion co-substitute for Ca<sup>2+</sup> ion on the A site of CaTiO<sub>3</sub>, results show that A site substitution at low amount is effective to form CaTiO<sub>3</sub> phase with perovskite structure. Mg and TiO<sub>2</sub> preferentially form perovskite structure to improve the sintering of CaTiO<sub>3</sub> ceramics, where the role of Mg is flux for whole sintering process. On the other hand, although unequal valence substitution introduces vacancy in structure, Nd<sup>3+</sup> substitution for Ca<sup>2+</sup> at suitable amount ameliorates the sintering of CaTiO<sub>3</sub>. The unequal valence substitution, whether on A site or B site is beneficial to sintering as reported by previous papers [11, 12]. We suppose that Nd<sup>3+</sup> ion addition introducing vacancy in structure and Mg<sup>2+</sup> ion addition bringing liquid sintering behavior to promote the dissolution of CaO and enhance the form of CaTiO<sub>3</sub> perovskite phase.

Figure 1 exhibits the microstructures of CaTiO<sub>3</sub> and modified ceramics by Ti, Nd, Mg and Li ions. The CaTiO<sub>3</sub> ceramic without additive has low density and shows different sintering mechanism from the modified sample's, where the sintering process of CaTiO<sub>3</sub> controlled by solid-state ion diffusion, the neck structure and pore between particles as well as the growth lines of particles coarsening are observed in Fig. 1(a), which means the particles form sintering bonds without densification even at high temperature more than that of modified samples. It seems that all samples with Ti, Nd, Mg and Li ion addition have relative density structures.

Table 1 demonstrates the relationship between additives amount and sintering properties of CaTiO<sub>3</sub>. As the substitution amount increasing, the secondary phases and sintering temperature of CaTiO<sub>3</sub> sample reduce. Nd and Mg ion addition effectively improve the sintering characteristics of CaTiO<sub>3</sub>, where the density increase from 3.96 to 4.42 g/cm<sup>3</sup> since the molecular weight of Nd is large than that of Ca. The perovskite structure

Table 1. The sintering properties of Ca<sub>1-y</sub>M<sub>2y/3</sub>TiO<sub>3</sub> ceramics.

Substitution amount	Sintering condition	Bulk density (g/cm <sup>3</sup> )	Secondary phases
0.0	1260°C/3 h	3.96	Dissociative CaO, TiO <sub>2</sub>
0.1	1260°C/3 h	3.99	Dissociative CaO, TiO <sub>2</sub>
0.2	1250°C/3 h	4.01	Dissociative TiO <sub>2</sub>
0.3	1220°C/3 h	4.16	Trace TiO <sub>2</sub>
0.33	1220°C/3 h	4.42	Trace TiO <sub>2</sub>
0.4	1220°C/3 h	4.42	Trace TiO <sub>2</sub>

and trace  $\text{TiO}_2$  phase are obtained at  $1220^\circ$  for 3 h for  $y \geq 0.3$  in  $\text{Ca}_{1-y}\text{M}_{2y/3}\text{TiO}_3$  ceramics.

In this paper, we used  $\text{Ca}_{0.67}\text{M}_{0.22}\text{TiO}_3$  ( $y = 0.33$ ) ceramics as a matrix (shown in Fig. 2:  $x = 0.0$ ) to investigate the sintering characteristic and di-

electric properties of  $\text{CaTiO}_3$  ceramics modified by  $\text{Li}_{1/2}\text{Nd}_{1/2}\text{TiO}_3$ . Figure 2 shows the XRD spectra of  $(1-x)\text{Ca}_{0.67}\text{M}_{0.22}\text{TiO}_3 - x\text{Li}_{1/2}\text{Nd}_{1/2}\text{TiO}_3$  ceramics sintered at different temperature. All samples are of perovskite structure, but with  $\text{Li}_{1/2}\text{Nd}_{1/2}\text{TiO}_3$

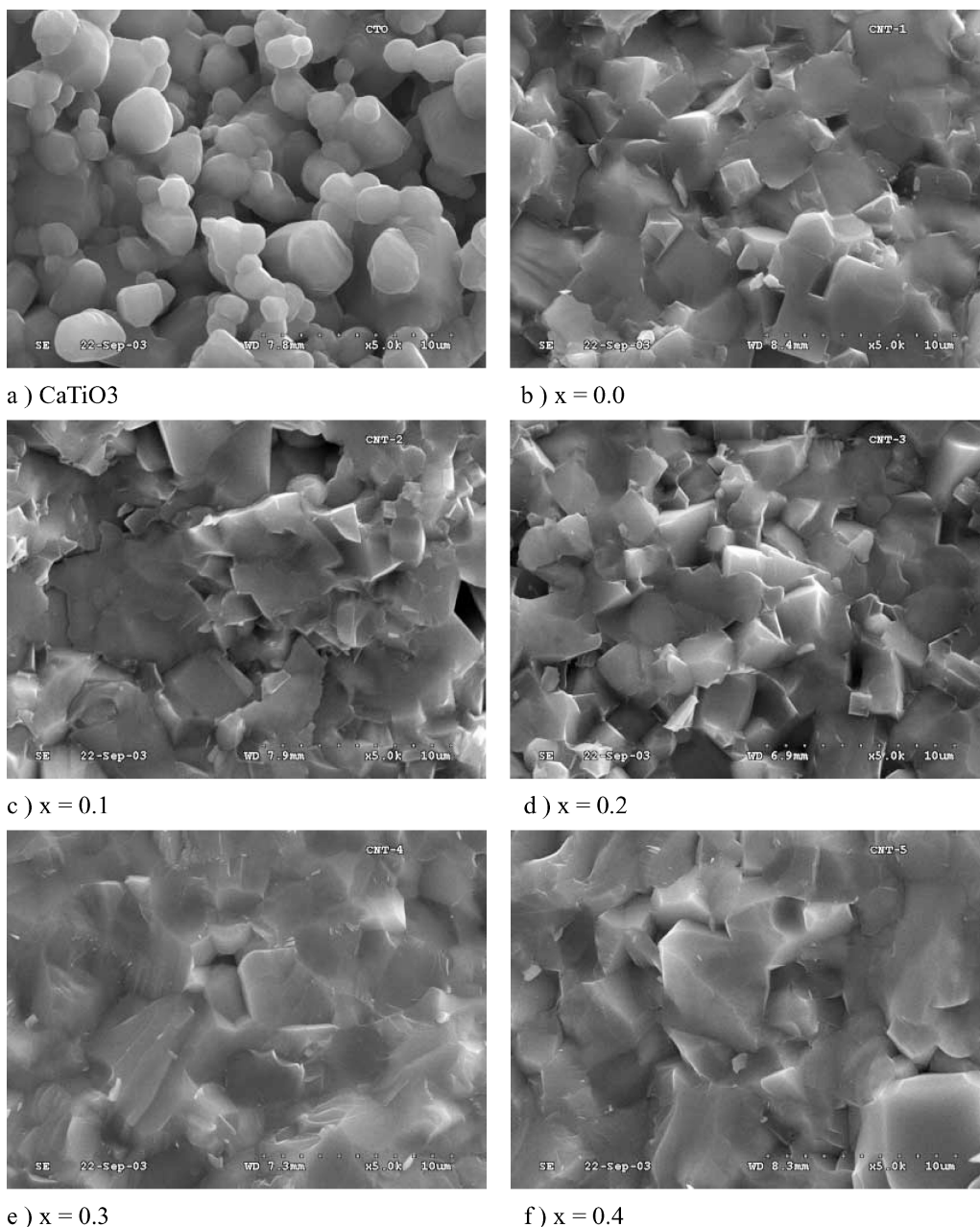


Fig. 1. Microstructures of  $\text{CaTiO}_3$  and  $(1-x)\text{Ca}_{0.67}\text{M}_{0.22}\text{TiO}_3 - x\text{Li}_{1/2}\text{Nd}_{1/2}\text{TiO}_3$  ceramics sintered at: (a)  $1260^\circ\text{C}$ ; (b)  $1220^\circ\text{C}$ ; (c)  $1180^\circ\text{C}$ ; (d)  $1180^\circ\text{C}$ ; (e)  $1200^\circ\text{C}$ ; (f)  $1220^\circ\text{C}$ ; (g)  $1220^\circ\text{C}$ ; (h)  $1220^\circ\text{C}$ ; (i)  $1220^\circ\text{C}$ ; and (j)  $1280^\circ\text{C}$ .  
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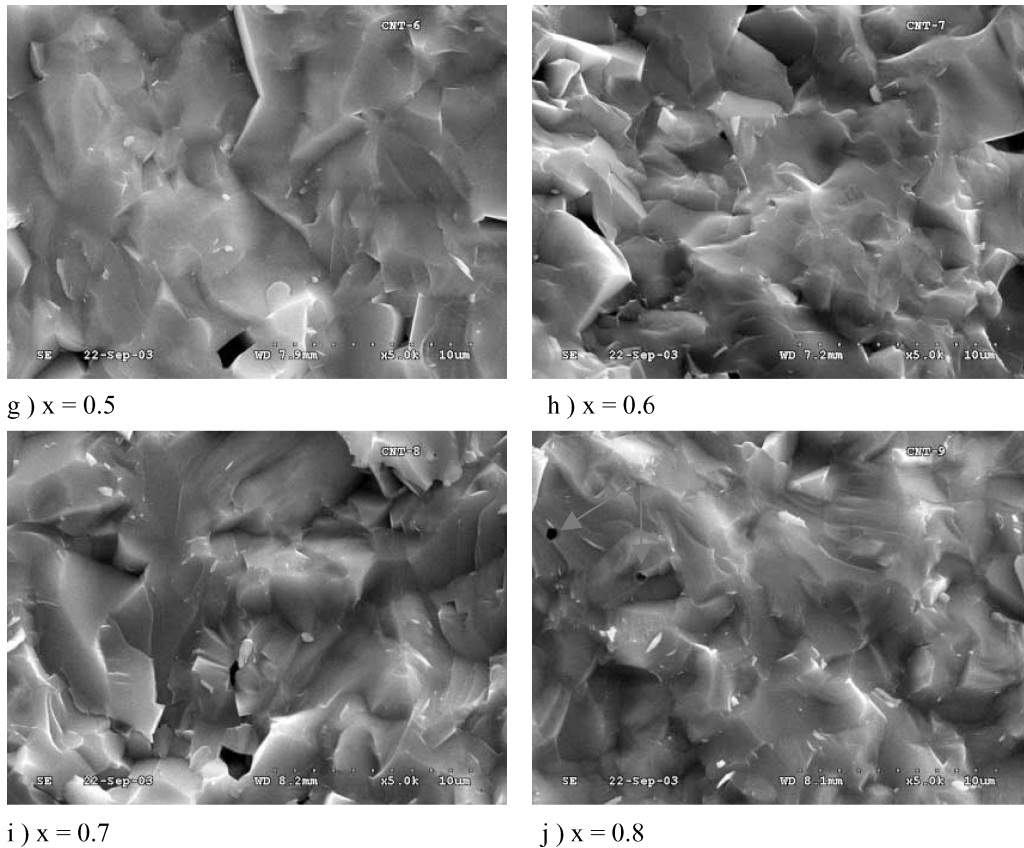


Fig. 1. (Continued).

addition the structure of  $\text{CaTiO}_3\text{-Li}_{1/2}\text{Nd}_{1/2}\text{TiO}_3$  binary system change from orthorhombic structure of  $\text{CaTiO}_3$  to tetragonal structure of  $\text{Li}_{1/2}\text{Nd}_{1/2}\text{TiO}_3$ . As is illustrated in Fig. 2, the orthorhombic structure exists in the composition range of  $x < 0.4$ , which is consistent with Kim's reports [7]. When  $x \geq 0.4$ , the superlattice peaks appear (shown by arrowhead), which means a tetragonal crystal structure of space group  $P\text{-}4\text{b}2$  where  $\text{Li}^{1+}$  and  $\text{Nd}^{3+}$  are ordered along the  $c$ -axis. Furthermore, the relative intensity of superlattice peaks becomes stronger as  $\text{Li}_{1/2}\text{Nd}_{1/2}\text{TiO}_3$  increasing.

In  $(1-x)\text{Ca}_{0.67}\text{M}_{0.22}\text{TiO}_3 - x\text{Li}_{1/2}\text{Nd}_{1/2}\text{TiO}_3$  binary system ceramics in this paper, the bulk density is sensitive to the structure change and densification. The density reduces with  $\text{Li}_{1/2}\text{Nd}_{1/2}\text{TiO}_3$  addition for  $x \leq 0.3$ , and increases up till to  $x = 0.7$ , then reduces again for  $x = 0.8$  (Fig. 3). For  $x < 0.4$ , the main phase has the orthorhombic structure of  $\text{Ca}_{0.67}\text{M}_{0.22}\text{TiO}_3$  with the end density of  $4.42 \text{ g/cm}^3$ , a small amount of

$\text{Li}_{1/2}\text{Nd}_{1/2}\text{TiO}_3$  addition contributes to the density decrease, but enhances the densification. As illustrated in Fig. 1, the porous structure clearly exists for  $x < 0.4$  samples and decreases as  $x$  increase. For  $x \geq 0.4$ , the density increases due to densification as demonstrated in Fig. 1 and the samples have the tetragonal structure of  $\text{Li}_{1/2}\text{Nd}_{1/2}\text{TiO}_3$  with end density of  $4.4 \text{ g/cm}^3$ , but it is unexplainable that the density increases from  $4.4$  to  $4.96 \text{ g/cm}^3$ . It must consider the effect of substitution amount of Nd on the density due to its large molecular weight, or, there are other compounds with high bulk density but the XRD spectra do not show. As for  $x = 0.8$ , the possibility of density decrease is sintering difficult of  $(1-x)\text{CaTiO}_3 - x\text{LiNdTiO}_3$ , where  $\text{Li}_{1/2}\text{Nd}_{1/2}\text{TiO}_3$  is the main component. In fact, the surface of sintering sample presents glass phase and pine pores, and whole sample shows bend shape due to asymmetrical shrinkage. Li evaporation in the surface is a possible reason at  $1280^\circ\text{C}$ . Another reason

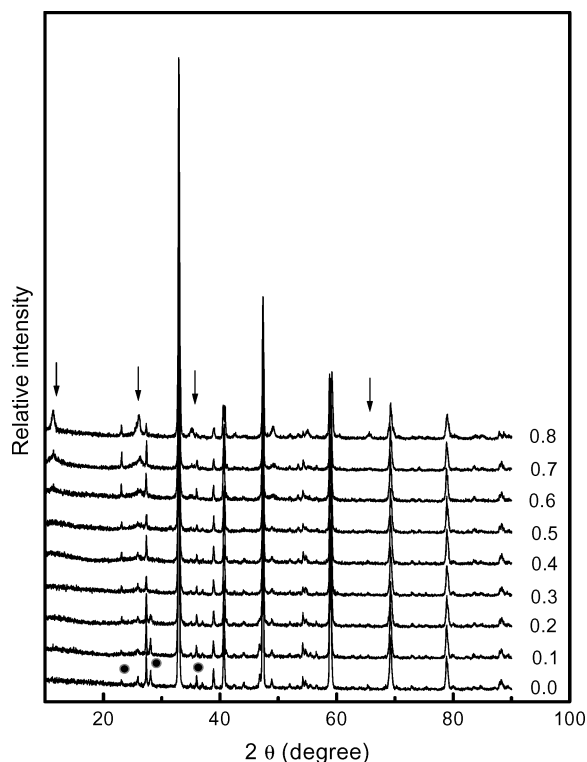


Fig. 2. The XRD spectra of  $(1-x)\text{Ca}_{0.67}\text{M}_{0.22}\text{TiO}_3 - x\text{Li}_{1/2}\text{Nd}_{1/2}\text{TiO}_3$  sintered ceramics as a function of  $x$ . (Arrow shows the superlattice phases, Circle denotes the second phases.)

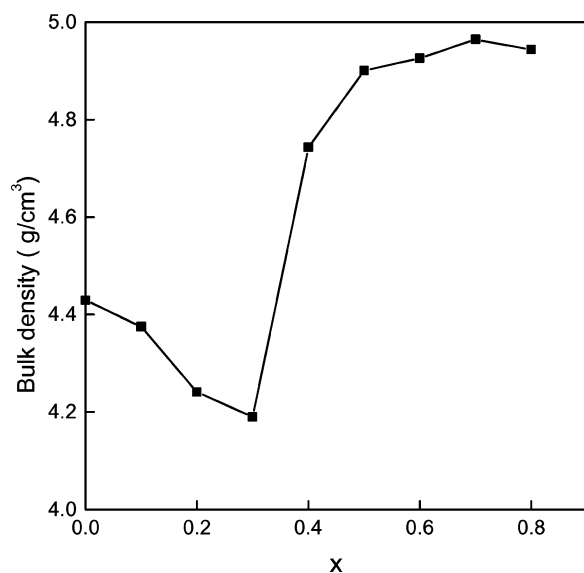


Fig. 3. Bulk density of  $(1-x)\text{Ca}_{0.67}\text{M}_{0.22}\text{TiO}_3 - x\text{Li}_{1/2}\text{Nd}_{1/2}\text{TiO}_3$  ceramics sintered at  $1220^\circ$  ( $x=0.8$  sintered at  $1280^\circ$ ) as a function of  $x$ .

maybe  $\text{Li}_{1/2}\text{Nd}_{1/2}\text{TiO}_3$  limited solubility into  $\text{CaTiO}_3$  contribute to component enrichment resulting in the local over sintering: the initial point contacts between particles grow into necks, subsequently, grain boundaries emerge and the pores form lenticular shapes that are deformed by grain boundary motion until pore boundary separation, where a spherical pore is left inside the grain (pointed by arrowhead in Fig. 1(j)). Furthermore,  $\text{Li}^+$  ion and  $\text{Nd}^{3+}$  ion arrange in local order accompanied with local composition change to form morphotropic phase boundary leading to densification difficult [13]. All factors contribute to bulk density decrease for  $x=0.8$  sample.

Figure 4 shows the frequency dependence of the room temperature dielectric constant of  $(1-x)\text{Ca}_{0.67}\text{M}_{0.22}\text{TiO}_3 - x\text{Li}_{1/2}\text{Nd}_{1/2}\text{TiO}_3$  ceramics as a function of  $x$ . The room temperature dielectric constant of the sample  $x=0.0$  is almost frequency independent.  $\text{Li}_{1/2}\text{Nd}_{1/2}\text{TiO}_3$  addition causes the small change of dielectric constant at low frequencies for  $x \leq 0.3$ , and great influence at all test frequencies for  $x > 0.4$ . This trend is contributed to the change of density and microstructure of samples. In general, polarization comprises two factors: relaxation polarization and resonance polarization, the former play main role at low frequencies, and respond to the massage of sample structure. The latter play important part

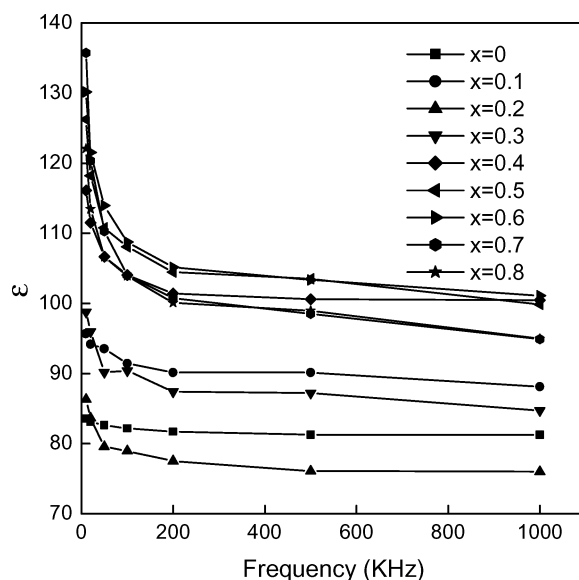


Fig. 4. Frequency dependence of the room temperature dielectric constant of  $(1-x)\text{Ca}_{0.67}\text{M}_{0.22}\text{TiO}_3 - x\text{Li}_{1/2}\text{Nd}_{1/2}\text{TiO}_3$  ceramics as a function of  $x$ .

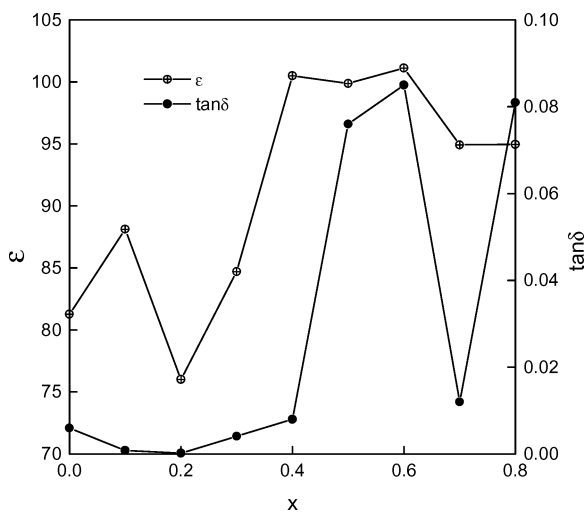


Fig. 5. Room temperature dielectric properties of  $(1-x)$ - $\text{Ca}_{0.67}\text{M}_{0.22}\text{TiO}_3 - x\text{Li}_{1/2}\text{Nd}_{1/2}\text{TiO}_3$  ceramics as a function of  $x$  (at 1 MHz).

at high frequencies. Microstructure difference, morphotropic phase boundary, pores, defect, etc. are the main resource of relaxation polarization, but they do not work at high frequencies. Thus,  $\text{Li}_{1/2}\text{Nd}_{1/2}\text{TiO}_3$  addition more than 40 mol% into  $\text{CaTiO}_3$  brings microstructure change to cause dielectric constant raise at low frequencies but little influence at high frequencies.

As the reason mentioned above, the dielectric loss of  $(1-x)\text{Ca}_{0.67}\text{M}_{0.22}\text{TiO}_3 - x\text{Li}_{1/2}\text{Nd}_{1/2}\text{TiO}_3$  ceramics has the same change trend (shown in Fig. 5). The dielectric loss is sensitive to microstructure, pores and grain boundary in samples. The dielectric loss decline as  $\text{Li}_{1/2}\text{Nd}_{1/2}\text{TiO}_3$  increasing within  $x < 0.3$ , then ascend rapidly for  $x \geq 0.4$ , which is consistent with the microstructure of samples as illustrated in Fig. 1, the most densification samples of  $x = 0.2-0.3$  have the lowest dielectric loss. On the other hand,  $\text{Li}_{1/2}\text{Nd}_{1/2}\text{TiO}_3$  addition increases the dielectric constant of samples at whole range of  $x$  except  $x = 0.2$ , due to the increasing amount of  $\text{Nd}^{3+}$  ion with large ion polarizability than  $\text{Li}^+$  and  $\text{Ca}^{2+}$  ions in the matrix [14].

Figure 6 illustrates the temperature characteristic of dielectric constant of  $(1-x)\text{Ca}_{0.67}\text{M}_{0.22}\text{TiO}_3 - x\text{Li}_{1/2}\text{Nd}_{1/2}\text{TiO}_3$  at 1 MHz.  $\text{Nd}^{3+}$  and  $\text{Mg}^{2+}$  substitution for  $\text{Ca}^{2+}$  have effectively adjusted the temperature coefficient of dielectric constant  $\tau_\epsilon$  of  $\text{CaTiO}_3$  matrix to a suitable level with the value changing from  $-1500 \text{ ppm}/^\circ\text{C}$  to  $+275 \text{ ppm}/^\circ\text{C}$ , and  $\text{Li}_{1/2}\text{Nd}_{1/2}\text{TiO}_3$

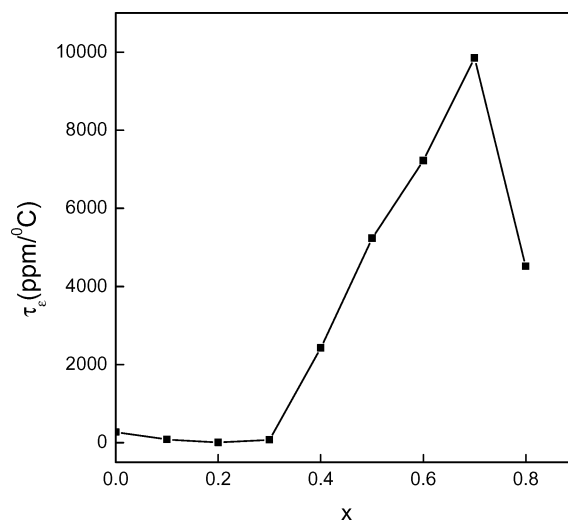


Fig. 6. Temperature coefficient of dielectric constant of  $(1-x)$ - $\text{Ca}_{0.67}\text{M}_{0.22}\text{TiO}_3 - x\text{Li}_{1/2}\text{Nd}_{1/2}\text{TiO}_3$  ceramics as a function of  $x$ .

improves  $\tau_\epsilon$  further for  $x \leq 0.3$ . With 20 mol%  $\text{Li}_{1/2}\text{Nd}_{1/2}\text{TiO}_3$  addition,  $\tau_\epsilon$  is modified to  $+7 \text{ ppm}/^\circ\text{C}$ , while that of the end member is  $+275 \text{ ppm}/^\circ\text{C}$ . As increasing  $\text{Li}_{1/2}\text{Nd}_{1/2}\text{TiO}_3$  addition,  $\tau_\epsilon$  increases rapidly in response to the crystal structure change for  $x \geq 0.4$  samples. The influence of microstructure and morphotropic phase boundary on the temperature coefficient of dielectric constant should be investigated further.

#### 4. Conclusion

$\text{Nd}^{3+}$  and  $\text{Mg}^{2+}$  co-substitution for  $\text{Ca}^{2+}$  on the A site can improve the sintering characteristic of  $\text{CaTiO}_3$  at low temperature. The structure of  $\text{CaTiO}_3$  change with the  $\text{Li}_{1/2}\text{Nd}_{1/2}\text{TiO}_3$  addition from orthorhombic to tetragonal, meanwhile, the dielectric constant increases, and the dielectric loss decrease at low addition amount then increase at high amount.  $\text{Li}_{1/2}\text{Nd}_{1/2}\text{TiO}_3$  addition makes the dielectric constant of  $\text{CaTiO}_3$  sensitive to the frequencies. Limited solubility of  $(\text{Li}_{1/2}\text{Nd}_{1/2})\text{TiO}_3$  in  $\text{CaTiO}_3$  promote the formation of a part solid solution compound to achieve the adjustment of  $\tau_\epsilon$  for  $\text{CaTiO}_3$ . The proper dielectric properties are obtained with  $\epsilon = 78$ ,  $\tan \delta = 0.0006$ ,  $\tau_\epsilon = +7 \text{ ppm}/^\circ\text{C}$  for  $0.8\text{Ca}_{0.67}(\text{Nd,Mg})_{0.22}\text{TiO}_3 - 0.2(\text{Li}_{1/2}\text{Nd}_{1/2})\text{TiO}_3$ , and  $\epsilon = 88$ ,  $\tan \delta = 0.0009$ ,  $\tau_\epsilon = +83 \text{ ppm}/^\circ\text{C}$  for  $0.9\text{Ca}_{0.67}(\text{Nd,Mg})_{0.22}\text{TiO}_3 - 0.1(\text{Li}_{1/2}\text{Nd}_{1/2})\text{TiO}_3$ .

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