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Ferroelectric-Dielectric Solid Solution and Composites for Tunable Microwave Application

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1. Introduction

Electric field tunable ferroelectric materials have attracted extensive attention in recent years due to their potential applications for tunable microwave device such as tunable filters, phased array antennas, delay lines and phase shifters (Maiti et al. 2007a; Rao et al. 1999; Romanofsky et al. 2000; Varadan et al 1992.; Zhi et al. 2002). $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$ and $\text{BaZr}_x\text{Ti}_{1-x}\text{O}_3$ have received the most attention due to their intrinsic high dielectric tunability. However, the high inherent materials loss and high dielectric constant has restricted its application in tunable microwave device. Various methods have been investigated to lower the dielectric constant and loss tangent of pure ferroelectrics.

Forming ferroelectric-dielectric composite is an efficient method to reduce material dielectric constant, loss tangent and maintain tunability at a sufficiently high level. For binary ferroelectric-dielectric composite (such as BST+MgO) (Chang & Sengupta 2002; Sengupta & Sengupta 1999), with the increase of dielectrics content, the dielectric constant and tunability of composites decrease. In order to decrease the dielectric constant of binary composite, it is necessary to increase the content of linear dielectric, and the tunability will decrease inevitably due to ferroelectric dilution. Replacing one dielectric by the combination of dielectrics with different dielectric constants and forming ternary ferroelectric-dielectric composite can decrease the dielectric constant of composite and maintain or even increase the tunability. This is beneficial for tunable application. The $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-Mg}_2\text{SiO}_4\text{-MgO}$ and $\text{BaZr}_{0.2}\text{Ti}_{0.8}\text{O}_3\text{-Mg}_2\text{SiO}_4\text{-MgO}$ composites exhibited relatively high tunability in combination with reduced dielectric permittivity and reduced loss tangent (He et al. 2010, 2011). With the increase of Mg_2SiO_4 content and the decrease of MgO content in $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-Mg}_2\text{SiO}_4\text{-MgO}$ composite, the dielectric constant decrease and the tunability remain almost unchanged. For $\text{BaZr}_{0.2}\text{Ti}_{0.8}\text{O}_3\text{-Mg}_2\text{SiO}_4\text{-MgO}$ composite, an anomalous relation between dielectric constant and tunability was observed: with the increase of Mg_2SiO_4 content (>30 wt%), the dielectric constant of composite decreases and the tunability increases. The anomalous increased tunability can be attributed to redistribution of the electric field. $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3\text{-Mg}_2\text{TiO}_4\text{-MgO}$ can also form ferroelectric ($\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$)-dielectric ($\text{Mg}_2\text{TiO}_4\text{-MgO}$) ternary composite and the dielectric constant can be decreased. With the increase of Mg_2TiO_4 content and the decrease of MgO content, the tunability of $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3\text{-Mg}_2\text{TiO}_4\text{-MgO}$ composite increase. The multiple-phase composites might complicate method to effectively deposit films, particularly if the dielectrics and ferroelectric are not compatible for simultaneous deposition or simultaneous adhesion with a substrate or with

each other. But ferroelectric-dielectric composite bulk ceramics show promising application, especially in accelerator: bulk ferroelectrics composites can be used as active elements of electrically controlled switches and phase shifters in pulse compressors or power distribution circuits of future linear colliders as well as tuning layers for the dielectric based accelerating structures (Kanareykin et al. 2006, 2009a, 2009b).

Forming ferroelectric-dielectric solid solution is another method to reduce material dielectric constant and loss tangent. Ferroelectric $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ can form solid solution with dielectrics $\text{Sr}(\text{Ga}_{0.5}\text{Ta}_{0.5})\text{O}_3$, $\text{La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$, $\text{La}(\text{Zn}_{0.5}\text{Ti}_{0.5})\text{O}_3$, and $\text{Nd}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ that have the same perovskite structure as the ferroelectrics (Xu et al. 2008, 2009). With the increase of the dielectrics content, the dielectric constant, loss tangent and tunability of solid solution decrease. $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ shows better dielectric properties than other solid solutions. Compared with ferroelectric-dielectric composite, forming solid solution can decrease the dielectric constant more rapidly when the doping content is nearly the same, and can also improve the loss tangent more effectively. On the other hand, ferroelectric-dielectric solid solution shows lower tunability than composites. The advantage of ferroelectric-dielectric solid solution is that single phase materials is favorable for the thin film deposition. The high dielectric field strength can be obtained easily in thin film to get high tunability.

In this chapter, we summarize the microstructures, dielectric tunable properties of ferroelectric-dielectric solid solution and composites, focusing mainly on our recent works.

2. Ferroelectric-dielectric composite

2.1 $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$ based composites

Various non-ferroelectric oxides, such as MgO , Al_2O_3 , ZrO_2 , Mg_2SiO_4 and MgTiO_3 , were added to $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$ to reduce the dielectric constant and loss tangent and maintain the tunability at sufficient high level (Chang & Sengupta 2002; Sengupta & Sengupta 1997, 1999). It is better that non-ferroelectric oxide doesn't react with ferroelectric $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$. MgO has low dielectric constant and loss tangent, can form ferroelectric ($\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$)-dielectric (MgO) composite. BST- MgO composite shows better dielectric properties. Mg_2SiO_4 is also a linear dielectrics with low dielectric constant, but it can react with $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$ to form $\text{Ba}_2(\text{TiO})(\text{Si}_2\text{O}_7)$, as shown in Fig. 1. For 10 mol% Mg_2SiO_4 mixed $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$, the major phase is $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$, and no Mg_2SiO_4 phase can be found except for two unidentified peaks at 27.6° and 29.7° (relative intensity: $\sim 1\%$). As the content of Mg_2SiO_4 increases from 20 to 60 mol%, the impurities phase of $\text{Ba}_2(\text{TiO})(\text{Si}_2\text{O}_7)$ is observed obviously and the relative content is increased with respect to the content of Mg_2SiO_4 . For 60 mol% Mg_2SiO_4 mixed $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ ceramics sintered at 1220°C , the strongest diffraction peak is the (211) face of $\text{Ba}_2(\text{TiO})(\text{Si}_2\text{O}_7)$ (not shown in Fig. 1). Therefore, for Mg_2SiO_4 added $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$, it is not as we expected that the ferroelectric ($\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$)-dielectric (Mg_2SiO_4) composite formed. The dielectric constants and unloaded Q values at microwave frequency were measured in the $\text{TE}_{01\delta}$ dielectric resonator mode using the Hakki and Coleman method by the network analyzer. Table 1 summarizes ϵ_r and the quality factor ($Q \times f = f_0 / \tan \delta$, where f_0 is the resonant frequency) at microwave frequencies for some $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-Mg}_2\text{SiO}_4$ ceramics. Increasing the Mg_2SiO_4 content results in a decrease of dielectric constant but has no obvious effect on the $Q \times f$ value. The low $Q \times f$ of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-Mg}_2\text{SiO}_4$ ceramics restricts their microwave application, and so the tunability has not been measured furthermore. The low $Q \times f$ is due to $\text{Ba}_2(\text{TiO})(\text{Si}_2\text{O}_7)$ which is a ferroelectrics with promising piezoelectric uses.

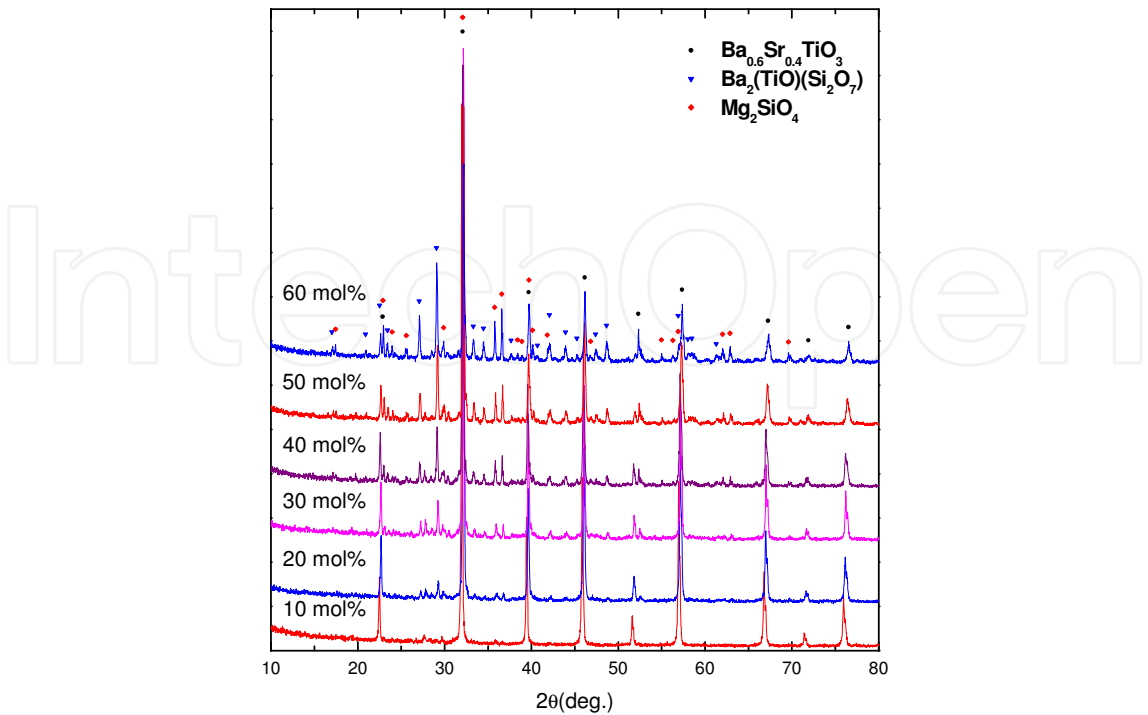


Fig. 1. The XRD patterns of Ba_{0.6}Sr_{0.4}TiO₃-Mg₂SiO₄ ceramics. The Mg₂SiO₄ content is 10-60mol%.

Mg ₂ SiO ₄ content (mol%)	Sintering temperature (°C)	f ₀ (GHz)	ε	tanδ	Q×f(GHz)
20	1260	1.79	683.7	0.016	112
40	1240	2.98	169.2	0.024	124

Table 1. Microwave dielectric properties of Ba_{0.6}Sr_{0.4}TiO₃-Mg₂SiO₄ ceramics

For Mg₂SiO₄-MgO added Ba_{0.6}Sr_{0.4}TiO₃, ferroelectric (Ba_{0.6}Sr_{0.4}TiO₃)-dielectric (Mg₂SiO₄-MgO) composite is formed, as shown in Fig. 2 (He et al., 2010). With the decrease of MgO content and the increase of Mg₂SiO₄ content, the diffraction peaks from MgO decrease gradually and the diffraction peaks from Mg₂SiO₄ increase. Therefore, Mg₂SiO₄-MgO combination can prohibit the formation of Ba₂(TiO)(Si₂O₇) phase.

Fig. 3 shows the FESEM images of Ba_{0.6}Sr_{0.4}TiO₃-Mg₂SiO₄-MgO composites sintered at 1350°C for 3h. The FESEM image and element mapping of 40Ba_{0.6}Sr_{0.4}TiO₃-12Ba_{0.6}Sr_{0.4}TiO₃-48MgO as determined by energy dispersive spectroscopy (EDS) are shown in Fig. 4. Three kind of different grains can be found clearly: light grains with average grain size of about 2μm, nearly round larger grains and dark grains with sharp corners. The element mapping of Si Kα1 and Ti Kα1 in Fig. 4 can show the distribution of Mg₂SiO₄ and Ba_{0.6}Sr_{0.4}TiO₃ grains clearly. Therefore, we can identify that light grains are Ba_{0.6}Sr_{0.4}TiO₃, the dark, larger grains are MgO, and dark grains with sharp corners are Mg₂SiO₄. With the decrease of MgO content and the increase of Mg₂SiO₄ content, more and more Mg₂SiO₄ grains with different size can be found (Fig. 4). It is consistent with the XRD results. We can conclude that Mg₂SiO₄ and MgO were randomly dispersed relative to ferroelectric Ba_{0.6}Sr_{0.4}TiO₃ phase.

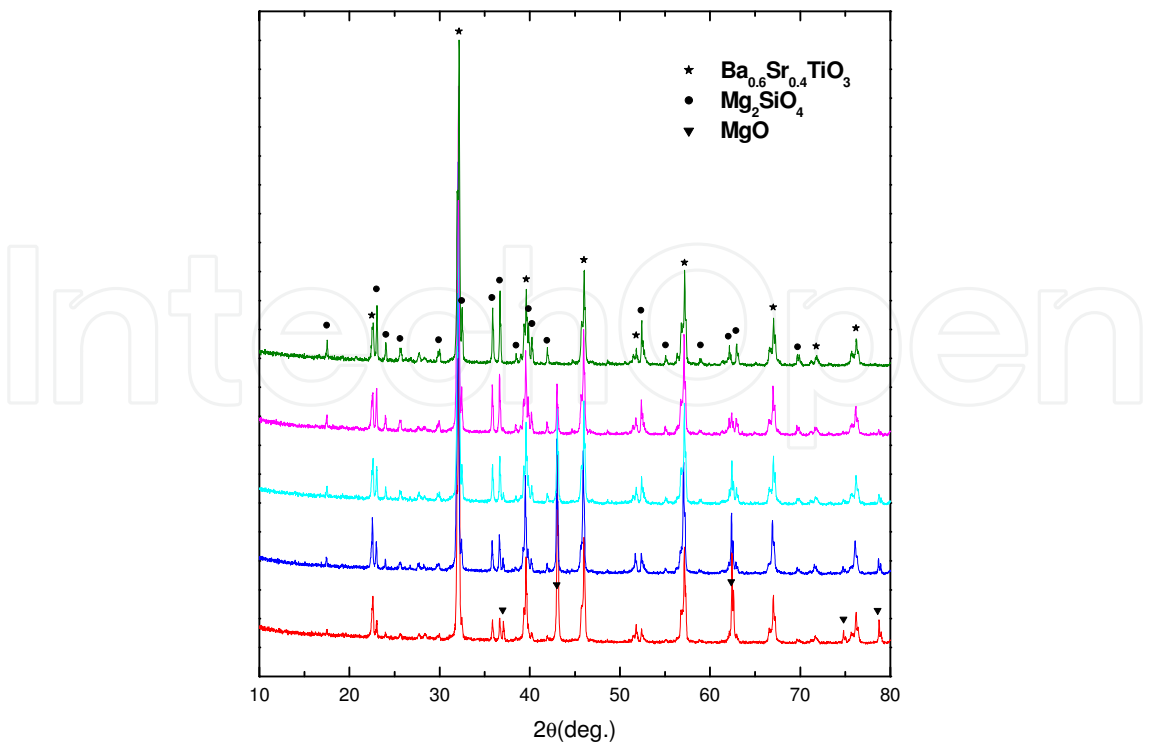


Fig. 2. The XRD patterns of 40Ba_{0.6}Sr_{0.4}TiO₃-60(Mg₂SiO₄-MgO) composite ceramics sintered at 1350°C for 3h. From bottom to top, the MgO content is 48 wt%, 36 wt%, 30 wt%, 24 wt% and 12 wt%, respectively.

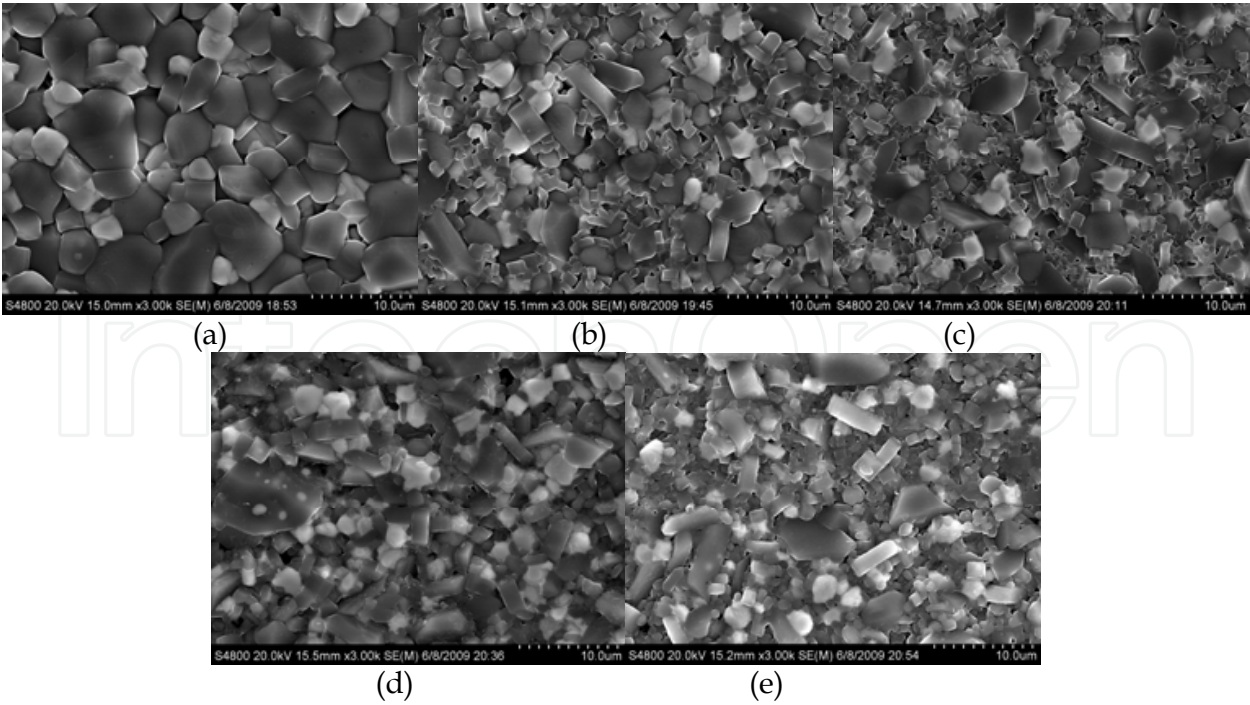


Fig. 3. FESEM images of 40Ba_{0.6}Sr_{0.4}TiO₃-60(Mg₂SiO₄-MgO) composites ceramics sintered at 1350°C for 3h. From (a) to (e), the MgO content is 48 wt%, 36 wt%, 30 wt%, 24 wt% and 12 wt%, respectively.

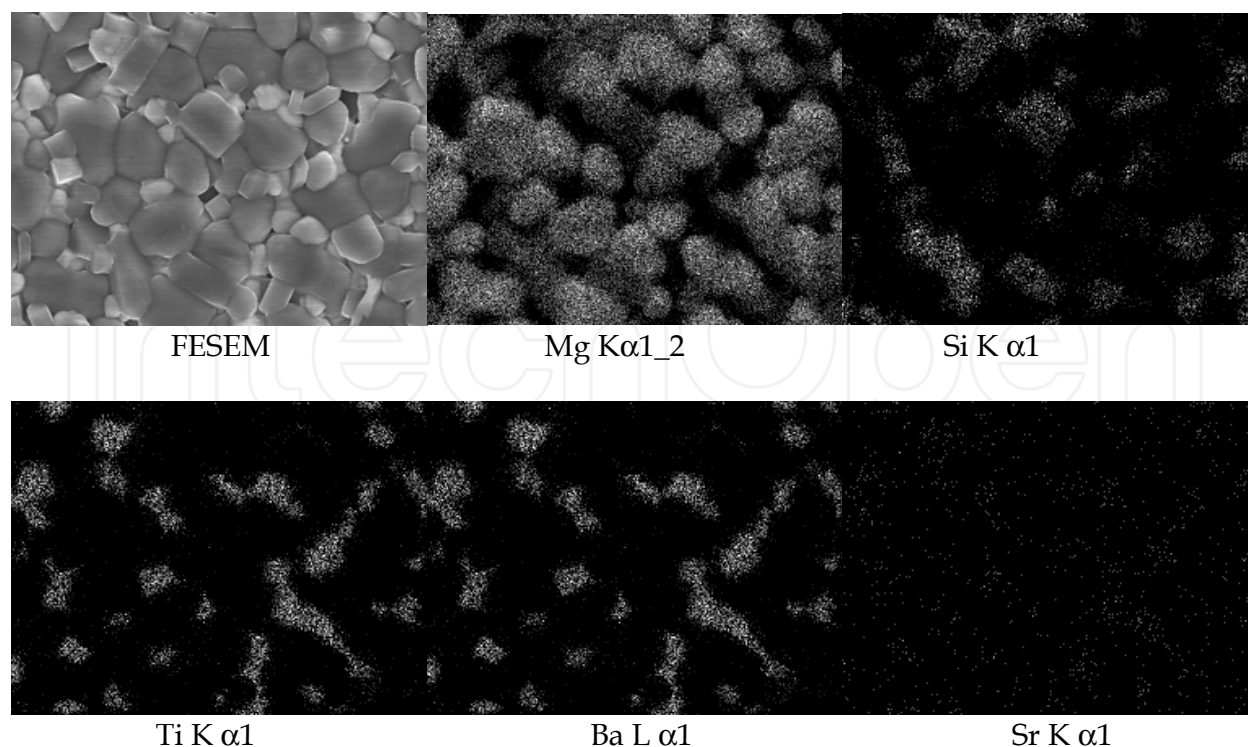


Fig. 4. FESEM image and element mapping of $40\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-}12\text{Mg}_2\text{SiO}_4\text{-}48\text{MgO}$ as determined by energy dispersive spectroscopy (EDS).

Because of the relatively low dielectric constant and loss tangent of Mg_2SiO_4 and MgO , it is expected that $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-Mg}_2\text{SiO}_4\text{-MgO}$ composites have lower dielectric constant and loss tangent. Fig. 5 shows the dielectric constant and loss tangent of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-Mg}_2\text{SiO}_4\text{-MgO}$ composite ceramics at 1MHz. The dielectric constant of composites is much smaller than that of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ ($\epsilon \sim 5160$ at 1MHz) (Chang & Sengupta, 2002; Sengptal & Sengupta 1999;). The loss tangent of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-Mg}_2\text{SiO}_4\text{-MgO}$ composites sintered at 1350°C is $\sim 0.0003\text{-}0.0006$, but the loss tangent of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ is ~ 0.0096 (Sengptal et al. 1999). Therefore, the composites have much smaller loss tangent than $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$.

The temperature dependence of dielectric properties for various $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-Mg}_2\text{SiO}_4\text{-MgO}$ composites (sintering temperature: 1350°C) measured at 100kHz is illustrated in Fig. 6. Broadened and suppressed dielectric peaks and shifts of Curie temperature T_c are observed. For $40\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-}12\text{Mg}_2\text{SiO}_4\text{-}48\text{MgO}$ ceramics, its ϵ_{\max} is ~ 176.5 at $T_c \sim 224\text{K}$. As the relative content of Mg_2SiO_4 increase, T_c is shifted slightly to lower temperatures, thus resulting in a decrease in dielectric constant at a given temperature; at the meantime, ϵ_{\max} decreases also. For $40\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-}30\text{Mg}_2\text{SiO}_4\text{-}30\text{MgO}$, ϵ_{\max} is ~ 140.1 at $\sim 216\text{K}$ and for $40\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-}48\text{Mg}_2\text{SiO}_4\text{-}12\text{MgO}$, ϵ_{\max} is ~ 126.8 at $\sim 214\text{K}$. With the decrease of temperature, the loss tangent increase.

Fig. 6 shows the effect of applied field on the tunability of the $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-Mg}_2\text{SiO}_4\text{-MgO}$ composites at 100kHz. The tunability of $40\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-}12\text{Mg}_2\text{SiO}_4\text{-}48\text{MgO}$ at 100kHz under at 2kV/mm is 10.5%. With the increase of Mg_2SiO_4 content, the tunability of $40\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-}24\text{Mg}_2\text{SiO}_4\text{-}36\text{MgO}$ decreases slightly to 9.2%. Further increasing Mg_2SiO_4 content results in a slight increase of tunability: $40\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-}48\text{Mg}_2\text{SiO}_4\text{-}12\text{MgO}$ composite has tunability of 10.2%.

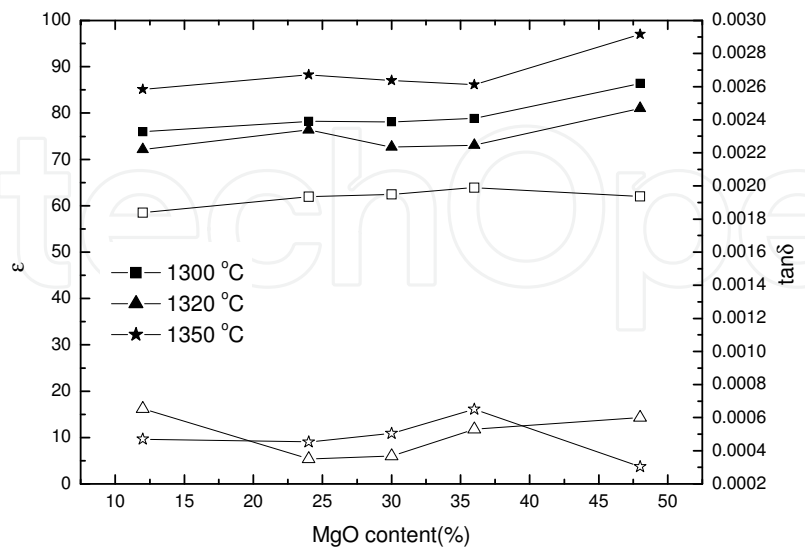


Fig. 5. Dielectric constant (solid) and loss tangent (open) of 40Ba_{0.6}Sr_{0.4}TiO₃-60(Mg₂SiO₄-MgO) composite ceramics sintered at different temperature (measure frequency: 1MHz).

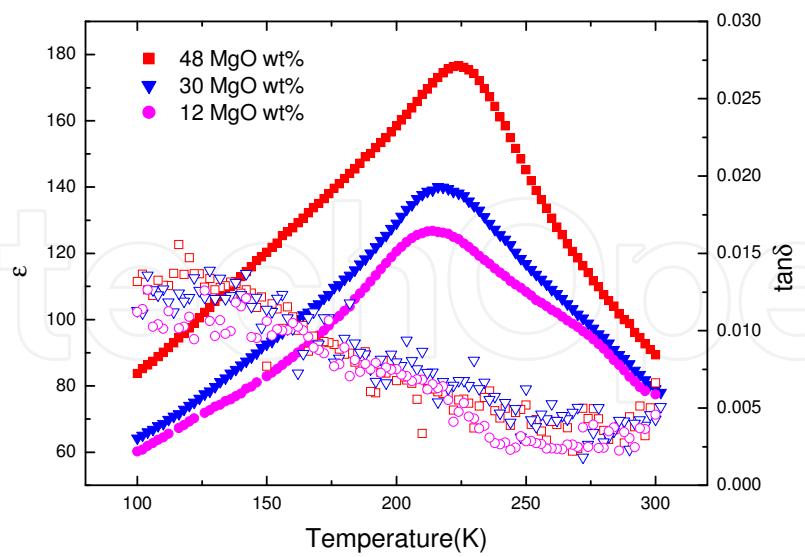


Fig. 6. Variation of dielectric constant (solid) and loss tangent (open) with temperature for 40Ba_{0.6}Sr_{0.4}TiO₃-60(Mg₂SiO₄-MgO) ceramics measured at 100kHz.

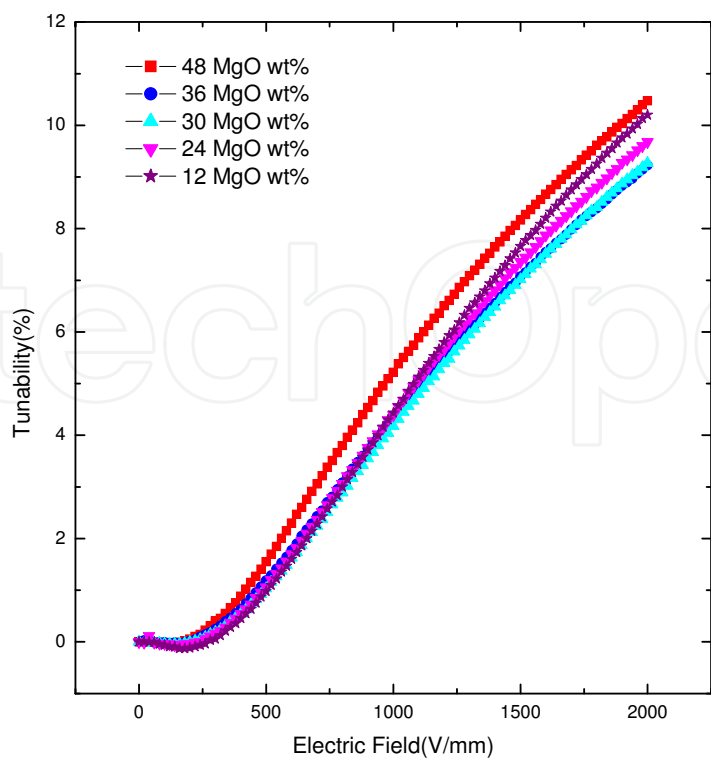


Fig. 7. The tunability of 40Ba_{0.6}Sr_{0.4}TiO₃-60(Mg₂SiO₄-MgO) composites at 100kHz (sintering temperature: 1350°C).

MgO content (wt.%)	f ₀ (GHz)	ε	tanδ	Q×f(GHz)
12	5.74	74.59	0.023	250
24	5.74	77.72	0.019	302
30	5.80	77.12	0.021	276
36	5.96	74.39	0.017	351
48	5.33	93.86	0.014	381

Table 2. Microwave Dielectric Properties of 40Ba_{0.6}Sr_{0.4}TiO₃-60(Mg₂SiO₄-MgO) ceramics

The room temperature microwave dielectric properties of 40Ba_{0.6}Sr_{0.4}TiO₃-60(Mg₂SiO₄-MgO) composites were summarized in Table 2. With the increase of Mg₂SiO₄ content, the dielectric constant remain almost the same and the Q×f value decrease.

Mg₂TiO₄ is a low loss tangent linear dielectrics and Mg₂TiO₄ added Ba_{1-x}Sr_xTiO₃ shows better tunable dielectric properties (Chou et al. 2007; Nenasheva et al. 2010). The XRD patterns of 40Ba_{0.6}Sr_{0.4}TiO₃-xMgO-(60-x)Mg₂TiO₄ (Fig. 8) show that ferroelectric (Ba_{0.6}Sr_{0.4}TiO₃)-dielectric (MgO-Mg₂TiO₄) composite is formed. On the other hand, impurity phase BaMg₆Ti₆O₁₉ is found in Mg₂TiO₄ doped Ba_{0.6}Sr_{0.4}TiO₃. The fomation of BaMg₆Ti₆O₁₉ depends on Ba/Sr ratio. BaMg₆Ti₆O₁₉ forms in Mg₂TiO₄ doped Ba_{0.6}Sr_{0.4}TiO₃ and Ba_{0.55}Sr_{0.45}TiO₃ but not Ba_{0.5}Sr_{0.5}TiO₃. Mg₂TiO₄-MgO combination can prohibit the formation of BaMg₆Ti₆O₁₉ phase. The FESEM images (Fig. 9) show clearly three kind of grains: Ba_{0.6}Sr_{0.4}TiO₃, Mg₂TiO₄ and MgO.

Table 3 shows the microwave dielectric properties of 40Ba_{0.6}Sr_{0.4}TiO₃-xMgO-(60-x)Mg₂TiO₄ ceramics. With the increase of MgO content, the dielectric constant decrease due to lower

dielectric constant of MgO. For $x=0-36$ wt%, the $Q \times f$ value remain unchanged. As a whole, the loss tangent is too high to be used for tunable microwave application.

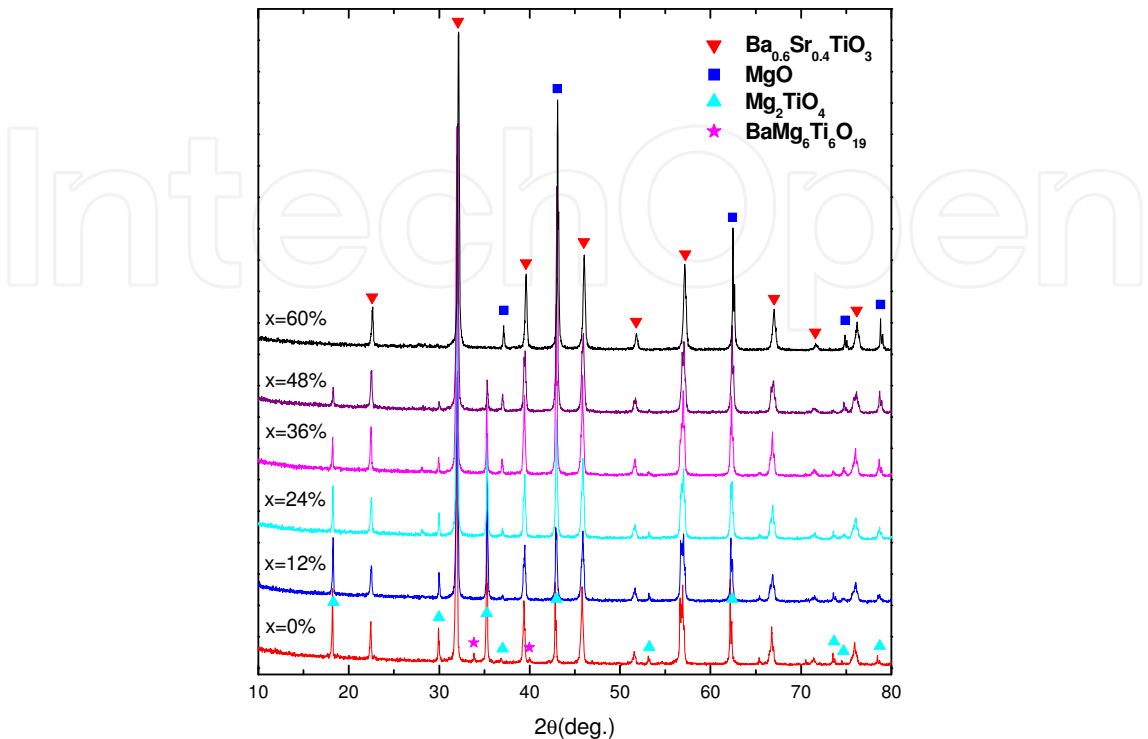


Fig. 8. The XRD patterns of $40\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3-x\text{MgO}-(60-x)\text{Mg}_2\text{TiO}_4$ ceramics

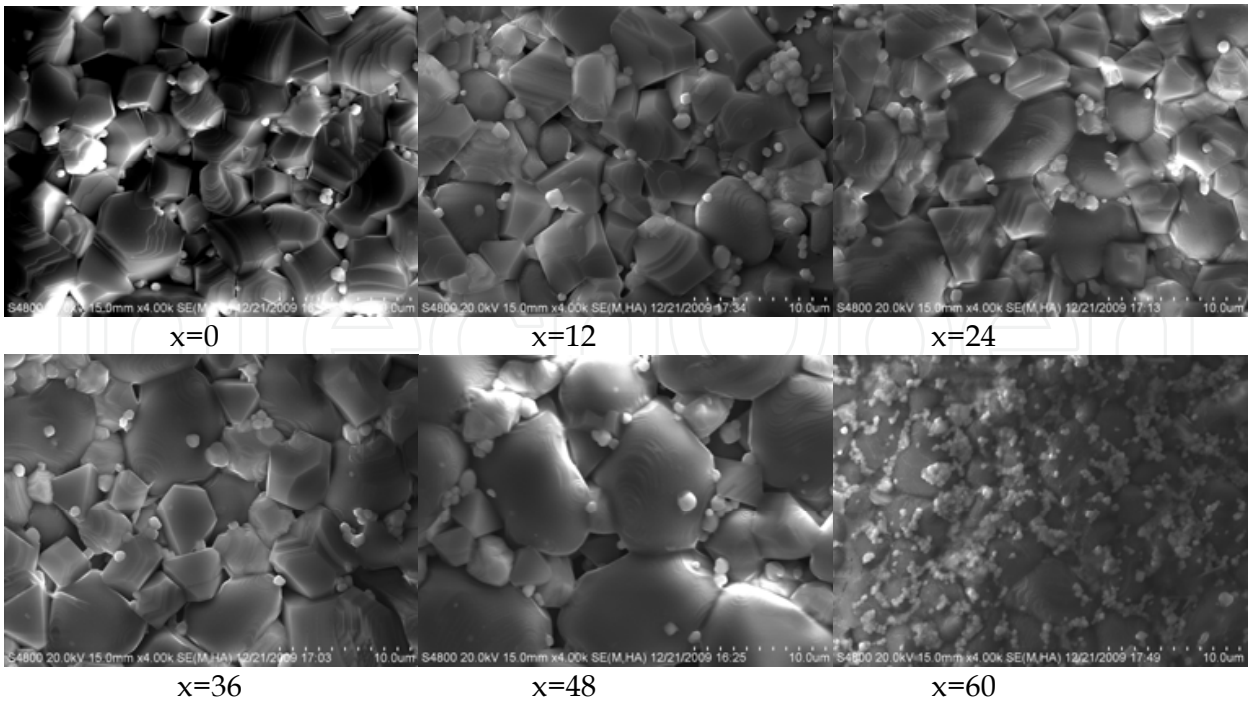


Fig. 9. FESEM images of $40\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3-x\text{MgO}-(60-x)\text{Mg}_2\text{TiO}_4$ composites ceramics sintered at 1400°C for 3h.

MgO content (wt.%)	f_0 (GHz)	ϵ	$\tan\delta$	$Q\times f$ (GHz)
0	2.83	193.40	0.034	83
12	2.67	226.76	0.034	79
24	2.96	220.25	0.035	85
36	3.00	207.66	0.036	83
48	3.53	199.71	0.034	104
60	4.80	109.63	0.013	369

Table 3. Microwave dielectric properties of $40\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-xMgO-(60-x)Mg}_2\text{TiO}_4$ ceramics

Increasing Sr/Ba ratio can decrease the dielectric constant and loss tangent of $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$. $40\text{Ba}_{0.5}\text{Sr}_{0.5}\text{TiO}_3\text{-xMgO-(60-x)Mg}_2\text{TiO}_4$ will has lower dielectric constant and loss tangent than $40\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-xMgO-(60-x)Mg}_2\text{TiO}_4$. We prepared $40\text{Ba}_{0.5}\text{Sr}_{0.5}\text{TiO}_3\text{-xMgO-(60-x)Mg}_2\text{TiO}_4$ ceramics and measured the tunability (Fig. 10). With the increase of Mg_2TiO_4 content, the tunability of composite increases. The tunability of $40\text{Ba}_{0.5}\text{Sr}_{0.5}\text{TiO}_3\text{-12MgO-48Mg}_2\text{TiO}_4$ is 16.6% at 2kV/mm and 28.5% at 3.9kV/mm, respectively. The corresponding value of $40\text{Ba}_{0.5}\text{Sr}_{0.5}\text{TiO}_3\text{-60Mg}_2\text{TiO}_4$ is 13.6% and 24.0% respectively. The higher tunability of $40\text{Ba}_{0.5}\text{Sr}_{0.5}\text{TiO}_3\text{-12MgO-48Mg}_2\text{TiO}_4$ is due to its higer dielectric constant ($\epsilon=150.2$) than $40\text{Ba}_{0.5}\text{Sr}_{0.5}\text{TiO}_3\text{-60Mg}_2\text{TiO}_4$ ($\epsilon=127.8$).

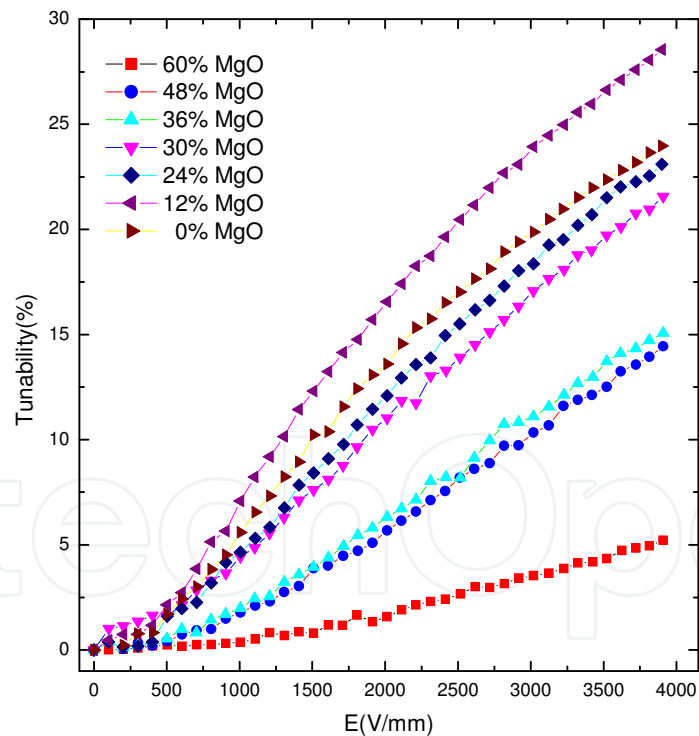


Fig. 10. The tunability of $40\text{Ba}_{0.5}\text{Sr}_{0.5}\text{TiO}_3\text{-xMgO-(60-x)Mg}_2\text{TiO}_4$ composites at 10kHz.

2.2 BaZrxTi1-xO3 based composites

BaZrxTi1-xO3 can form ferroelectric-dielectric composite with MgO (Maiti et al. 2007b, 2007c, 2008). High tunability and low loss tangent of the BaZrxTi1-xO3: MgO composites are

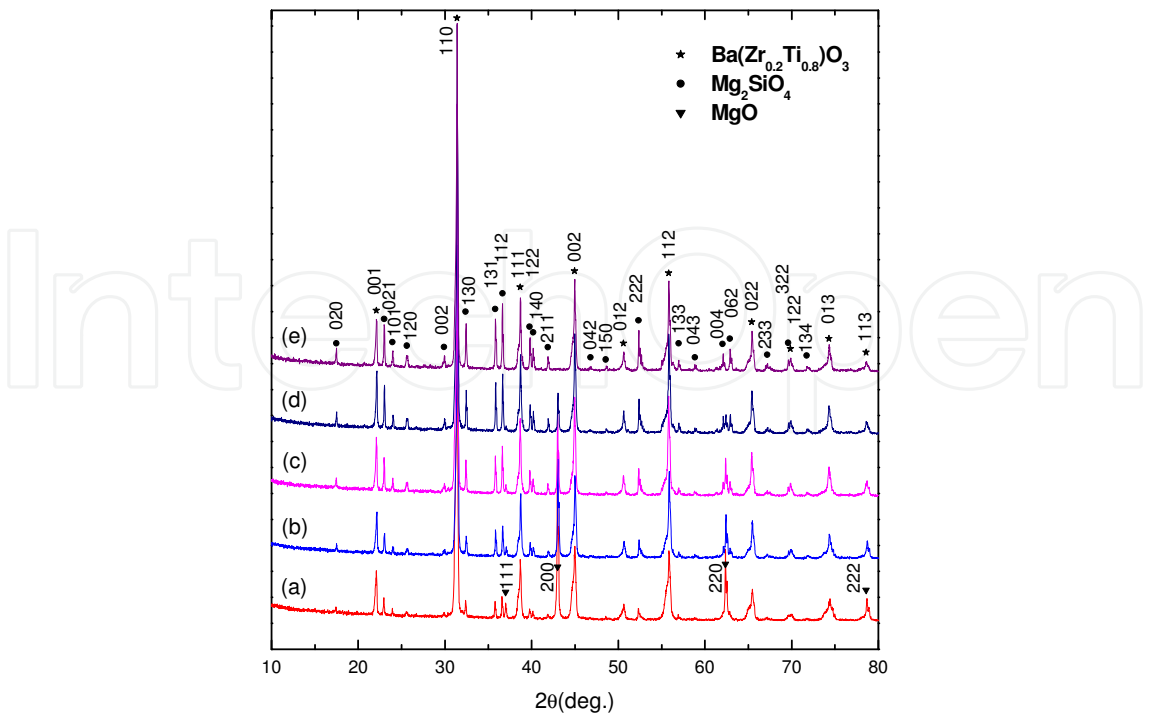


Fig. 11. The XRD patterns of $40\text{BaZr}_{0.2}\text{Ti}_{0.8}\text{O}_3$ -($60-x$) Mg_2SiO_4 - $x\text{MgO}$ composites ceramics sintered at 1350°C for 3h. (a) $x=48\text{wt}\%$, (b) $x=36\text{wt}\%$, (c) $x=30\text{wt}\%$, (d) $x=24\text{wt}\%$, (e) $x=12\text{wt}\%$.

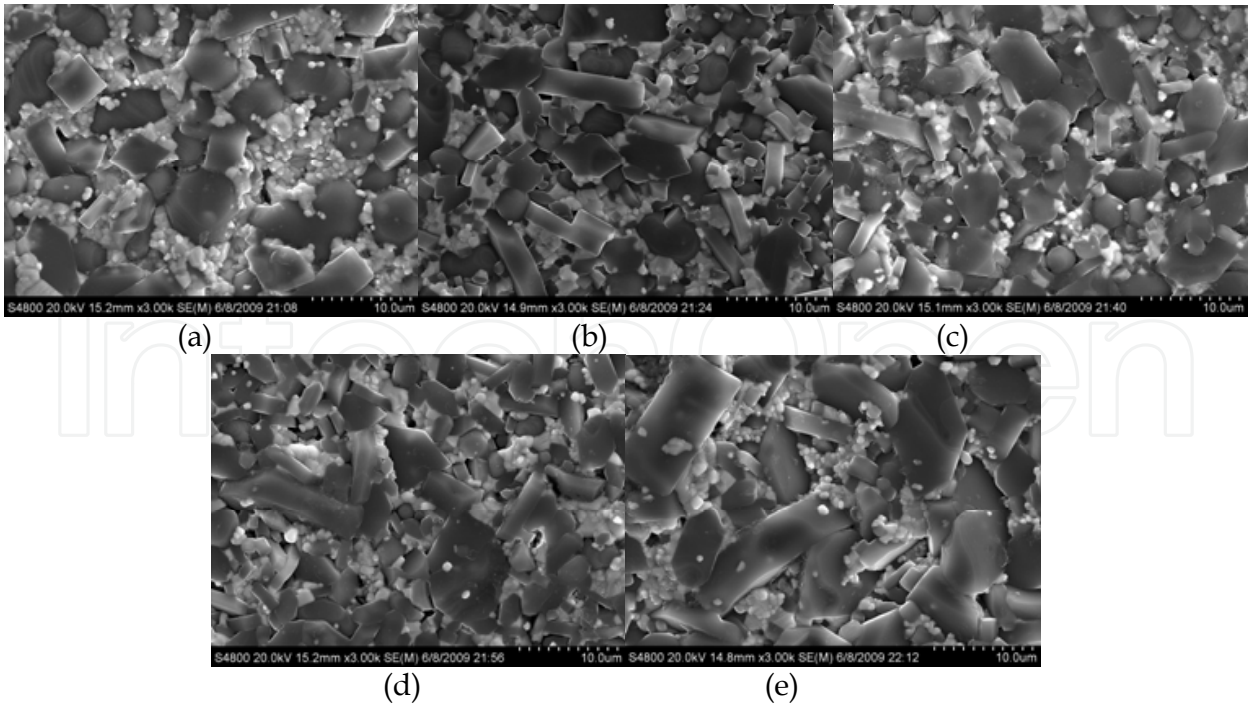


Fig. 12. FESEM images of $40\text{BaZr}_{0.2}\text{Ti}_{0.8}\text{O}_3$ -($60-x$) Mg_2SiO_4 - $x\text{MgO}$ composites ceramics sintered at 1350°C for 3h. From (a) to (e), $x=48\text{ wt}\%$, $36\text{ wt}\%$, $30\text{ wt}\%$, $24\text{ wt}\%$ and $12\text{ wt}\%$, respectively.

reported, but the sintering temperature is as high as 1500°C. We prepared BaZr_{0.2}Ti_{0.8}O₃-Mg₂SiO₄-MgO composite ceramics at 1350°C (He et al. 2011). The formation of ferroelectric (BaZr_{0.2}Ti_{0.8}O₃)-dielectric (Mg₂SiO₄-MgO) composite was proved by XRD patterns (Fig. 11). Similar to Ba_{0.6}Sr_{0.4}TiO₃-Mg₂SiO₄-MgO composites, three kind of grains: BaZr_{0.2}Ti_{0.8}O₃, Mg₂SiO₄ and MgO, can be identified (Fig. 12 and Fig. 13).

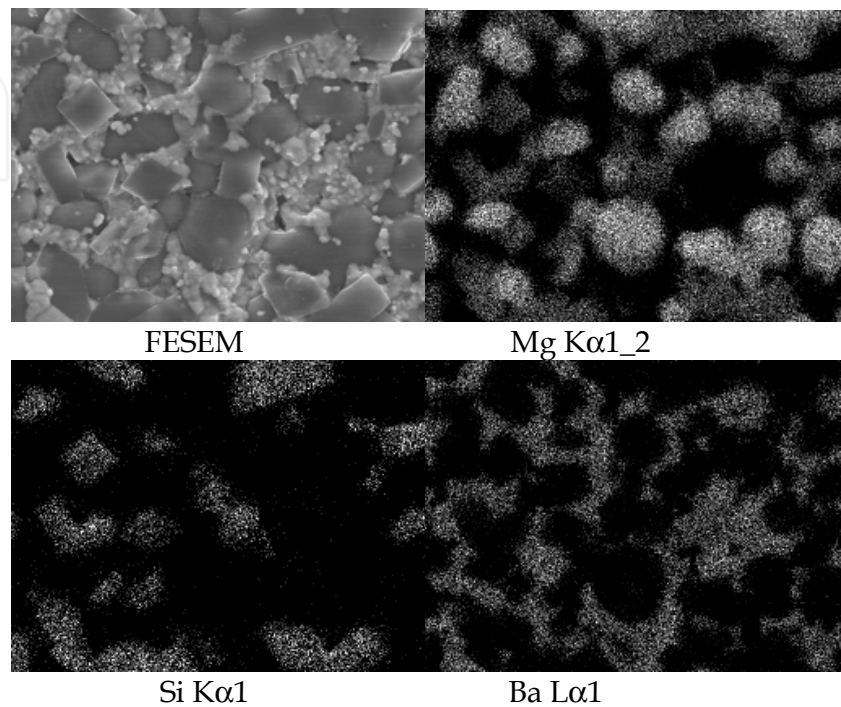


Fig. 13. FESEM image and element mapping of 40BaZr_{0.2}Ti_{0.8}O₃-12Mg₂SiO₄-48MgO as determined by energy dispersive spectroscopy (EDS).

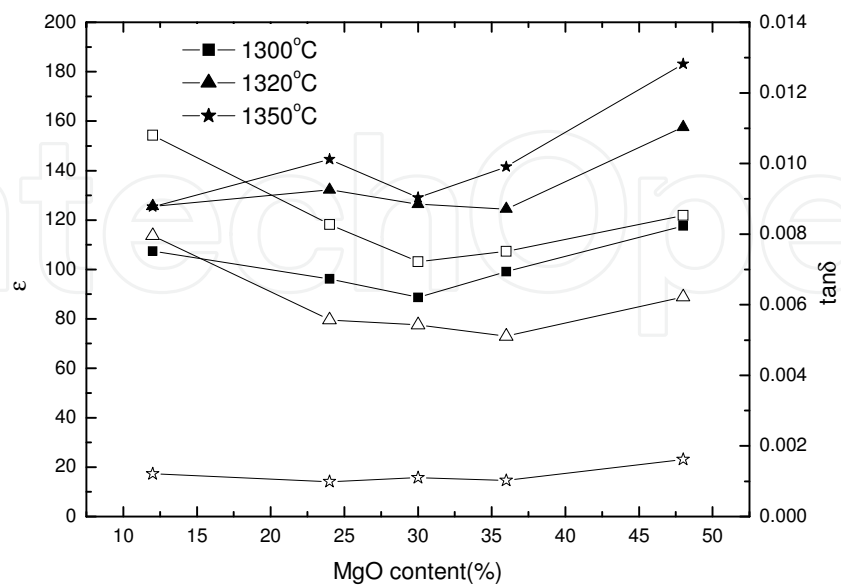


Fig. 14. Dielectric constant (solid) and loss tangent (open) of 40BaZr_{0.2}Ti_{0.8}O₃-(60-x)Mg₂SiO₄-xMgO composites ceramics sintered at various temperature (measure frequency: 1MHz).

Fig. 14 shows the dielectric constant and loss tangent of $\text{BaZr}_{0.2}\text{Ti}_{0.8}\text{O}_3\text{-Mg}_2\text{SiO}_4\text{-MgO}$ composite ceramics at 1MHz. With the increase of sintering temperature from 1300°C to 1350°C, the dielectric constant of the composites increase and the loss tangent decrease.

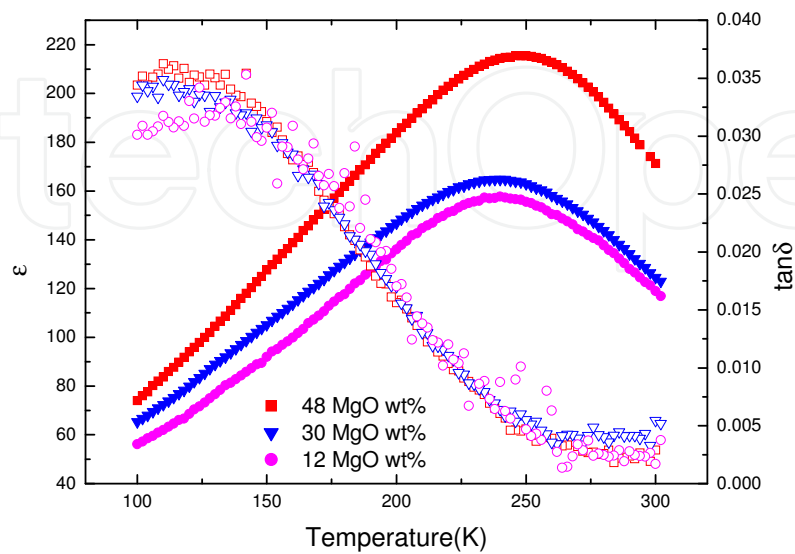


Fig. 15. Variation of dielectric constant (solid) and loss tangent (open) with temperature for $40\text{BaZr}_{0.2}\text{Ti}_{0.8}\text{O}_3\text{-(60-x)Mg}_2\text{SiO}_4\text{-xMgO}$ ceramics (sintering temperature: 1350°C) measured at 100kHz.

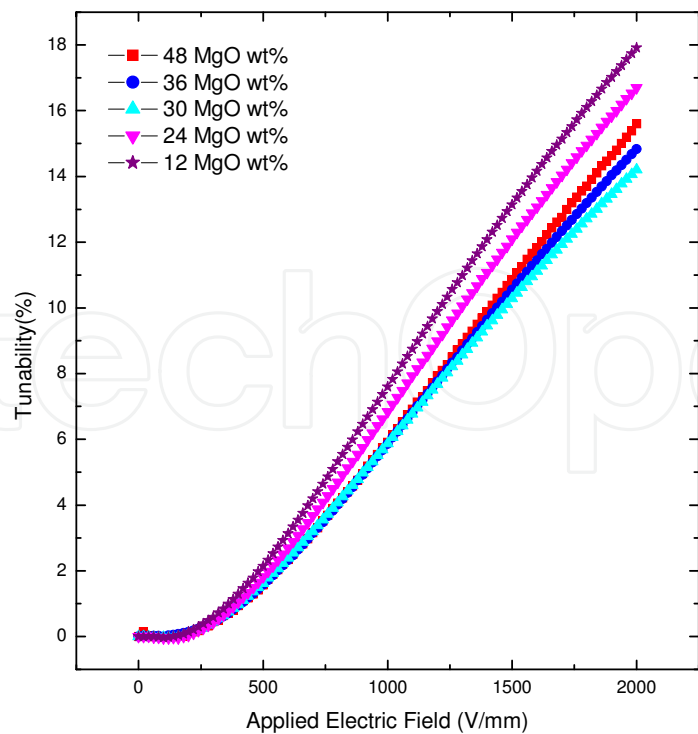


Fig. 16. The tunability of $40\text{BaZr}_{0.2}\text{Ti}_{0.8}\text{O}_3\text{-(60-x)Mg}_2\text{SiO}_4\text{-xMgO}$ composite ceramics at 100kHz at room temperature (sintering temperature: 1350°C).

Increasing Mg_2SiO_4 content tends to decrease the dielectric constant of composites. The dielectric constant and loss tangent of composite sintered at 1350°C is ~ 125 - 183 and ~ 0.0010 - 0.0016 , respectively, which is smaller than that of $\text{BaZr}_{0.2}\text{Ti}_{0.8}\text{O}_3$ (Maiti et al. 2007b).

The temperature dependence of dielectric properties for $\text{BaZr}_{0.2}\text{Ti}_{0.8}\text{O}_3$ - Mg_2SiO_4 - MgO composites (sintering temperature: 1350°C) measured at 100kHz is illustrated in Fig. 15. Compared with pure $\text{BaZr}_{0.2}\text{Ti}_{0.8}\text{O}_3$ bulk ceramic (Maiti et al. 2007b), broadened and suppressed dielectric peaks and shifts of Curie temperature T_c are observed with the addition of Mg_2SiO_4 and MgO . The results are similar to that of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ - Mg_2SiO_4 - MgO . For $40\text{BaZr}_{0.2}\text{Ti}_{0.8}\text{O}_3$ - $12\text{Mg}_2\text{SiO}_4$ - 48MgO ceramics, its ϵ_{max} decreases to ~ 215.5 and T_c shifts to lower temperature $\sim 246\text{K}$. For $40\text{BaZr}_{0.2}\text{Ti}_{0.8}\text{O}_3$ - $48\text{Mg}_2\text{SiO}_4$ - 12MgO , ϵ_{max} is ~ 157.7 at $\sim 240\text{K}$.

Fig. 16. shows the tunability of the $\text{BaZr}_{0.2}\text{Ti}_{0.8}\text{O}_3$ - Mg_2SiO_4 - MgO composites at 100kHz at room temperature. The tunability of $40\text{BaZr}_{0.2}\text{Ti}_{0.8}\text{O}_3$ - $12\text{Mg}_2\text{SiO}_4$ - 48MgO under 2kV/mm is 15.6% . With the increase of Mg_2SiO_4 content, the tunability of $40\text{BaZr}_{0.2}\text{Ti}_{0.8}\text{O}_3$ - $30\text{Mg}_2\text{SiO}_4$ - 30MgO decreases slightly to 14.2% . Further increasing Mg_2SiO_4 content results in an anomalous increase of tunability: $40\text{BaZr}_{0.2}\text{Ti}_{0.8}\text{O}_3$ - $48\text{Mg}_2\text{SiO}_4$ - 12MgO composite has lower dielectric constant than $40\text{BaZr}_{0.2}\text{Ti}_{0.8}\text{O}_3$ - $12\text{Mg}_2\text{SiO}_4$ - 48MgO but slightly higher tunability (17.9%).

3. Ferroelectric-dielectric solid solution

Forming ferroelectric-dielectric solid solution is another method to reduce material dielectric constant and loss tangent. Some non-ferroelectric complex oxides with perovskite structures have relatively low dielectric constant and low loss tangent. It is expected that they can be combined with barium strontium titanate to reduce material dielectric constant and loss tangent. Furthermore, it is possible for them to form solid solutions with barium strontium titanate because they have the same perovskite structure as barium strontium titanate. Single phase material is favorable for the thin film deposition. On the other hand, some perovskite oxide has positive temperature coefficient of dielectric constant and it can decrease the temperature coefficient of dielectric constant of barium strontium titanate above Curie temperature.

3.1 $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ - $\text{Sr}(\text{Ga}_{0.5}\text{Ta}_{0.5})\text{O}_3$ solid solution

$\text{Sr}(\text{Ga}_{0.5}\text{Ta}_{0.5})\text{O}_3$ has a comparatively small dielectric constant (27 at 1MHz), a positive temperature coefficient of dielectric constant (120ppmK^{-1}) and a low dielectric loss ($Q=8600$ at 10.6GHz) (Takahashi et al. 1997). The lattice constant ($a=0.3949\text{nm}$) of cubic perovskite structure $\text{Sr}(\text{Ga}_{0.5}\text{Ta}_{0.5})\text{O}_3$ is very close to that of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ ($a=0.3965\text{nm}$). Therefore, $\text{Sr}(\text{Ga}_{0.5}\text{Ta}_{0.5})\text{O}_3$ will be possible to form solid solution with $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ and reduce the dielectric constant of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$. The XRD results (Fig. 17.) prove that solid solution can be formed between $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ and $\text{Sr}(\text{Ga}_{0.5}\text{Ta}_{0.5})\text{O}_3$ under the preparative conditions (Xu et al. 2008).

Fig.18 shows the FESEM images of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ - $\text{Sr}(\text{Ga}_{0.5}\text{Ta}_{0.5})\text{O}_3$ ceramics sintered at 1600°C for 3h . The effect of $\text{Sr}(\text{Ga}_{0.5}\text{Ta}_{0.5})\text{O}_3$ content on the average grain size is not very obvious. We can also see that $0.9\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ - $0.1\text{Sr}(\text{Ga}_{0.5}\text{Ta}_{0.5})\text{O}_3$ has higher porosity than other compositions. The morphology of $0.5\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ - $0.5\text{Sr}(\text{Ga}_{0.5}\text{Ta}_{0.5})\text{O}_3$ shows difference from that of other three compositions.

The temperature dependence of dielectric properties for various $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ -

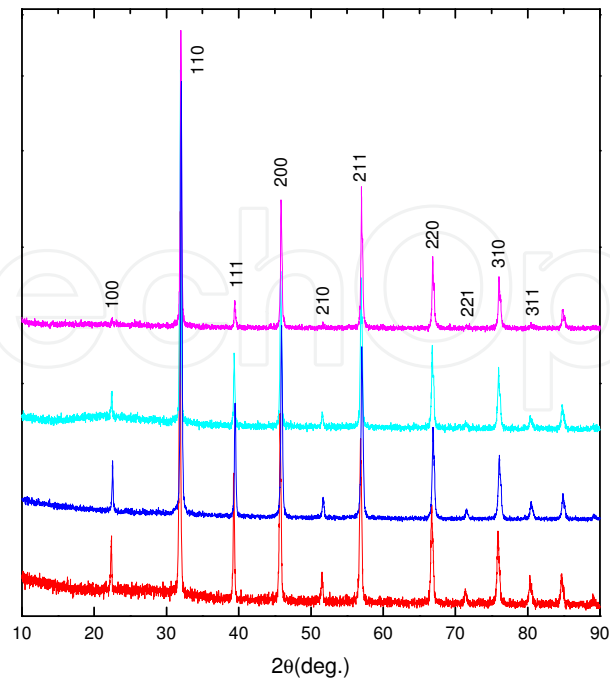


Fig. 17. The XRD patterns of Ba_{0.6}Sr_{0.4}TiO₃-Sr(Ga_{0.5}Ta_{0.5})O₃ ceramics sintered at 1600°C for 3h. From bottom to top, the Sr(Ga_{0.5}Ta_{0.5})O₃ content is 10, 20, 30 and 50mol%, respectively. The intensity is plotted on a log scale.

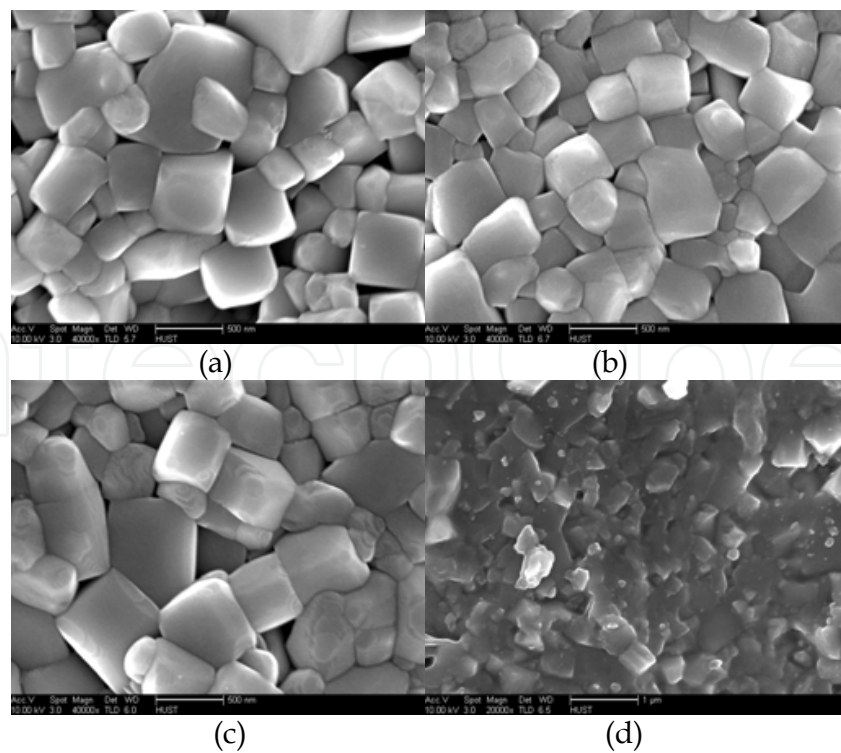


Fig. 18. FESEM images of Ba_{0.6}Sr_{0.4}TiO₃-Sr(Ga_{0.5}Ta_{0.5})O₃ ceramics sintered at 1600°C for 3h. From (a) to (d), the Sr(Ga_{0.5}Ta_{0.5})O₃ content is 10, 20, 30 and 50mol%, respectively.

Sr(Ga_{0.5}Ta_{0.5})O₃ ceramics (sintering temperature: 1600°C) measured at 100kHz is illustrated in Fig. 19. Broadened and suppressed dielectric peaks and shifts of Curie temperature T_C are observed with the addition of Sr(Ga_{0.5}Ta_{0.5})O₃. For 0.9Ba_{0.6}Sr_{0.4}TiO₃-0.1Sr(Ga_{0.5}Ta_{0.5})O₃ ceramics, its ε_{max} decreases to ~ 686 and T_c shifts to lower temperature ~250K. As more Sr(Ga_{0.5}Ta_{0.5})O₃ is added to Ba_{0.6}Sr_{0.4}TiO₃, T_c shifts to lower temperatures, thus resulting in a decrease in dielectric constant at a given temperature and ε_{max}. For 0.8Ba_{0.6}Sr_{0.4}TiO₃-0.2Sr(Ga_{0.5}Ta_{0.5})O₃, ε_{max} is ~335 at ~200K and for 0.5Ba_{0.6}Sr_{0.4}TiO₃-0.5Sr(Ga_{0.5}Ta_{0.5})O₃, ε_{max} is ~95 at ~100K. On the other hand, loss tangent increases on cooling. For 0.9Ba_{0.6}Sr_{0.4}TiO₃-0.1Sr(Ga_{0.5}Ta_{0.5})O₃ ceramics, there is small peak around ~250K. The loss tangent of 0.5Ba_{0.6}Sr_{0.4}TiO₃-0.5Sr(Ga_{0.5}Ta_{0.5})O₃ ceramics (not shown) is almost independent on temperature and fluctuates around 0.004 at the temperature range of 60K-300K.

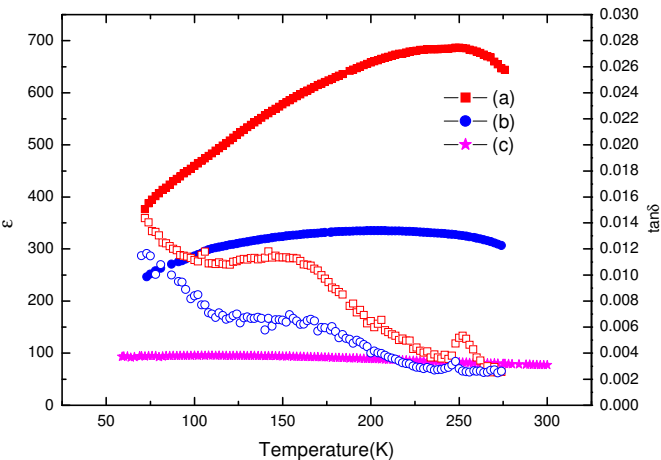


Fig. 19. Variation of dielectric constant (solid) and loss tangent (open) with temperature for Ba_{0.6}Sr_{0.4}TiO₃-Sr(Ga_{0.5}Ta_{0.5})O₃ ceramics (sintering temperature: 1600°C) measured at 100kHz: From (a) to (c), the Sr(Ga_{0.5}Ta_{0.5})O₃ content is 10, 20, and 50 mol%, respectively.

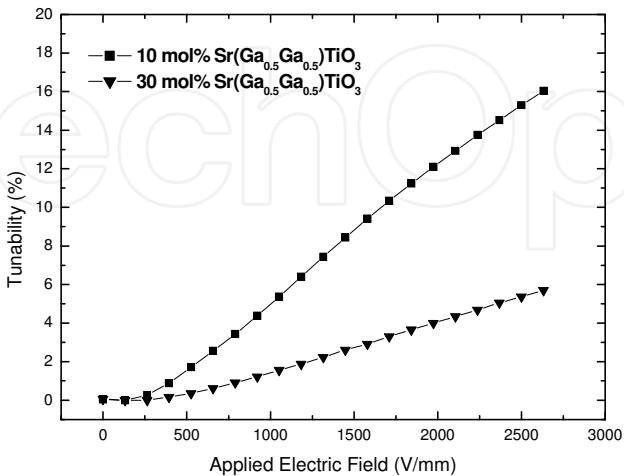


Fig. 20. The tunability of 0.9Ba_{0.6}Sr_{0.4}TiO₃-0.1Sr(Ga_{0.5}Ta_{0.5})O₃ and 0.7Ba_{0.6}Sr_{0.4}TiO₃-0.3Sr(Ga_{0.5}Ta_{0.5})O₃ at 100 kHz (sintering temperature: 1600°C).

Fig. 20 shows the tunability of the $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-Sr}(\text{Ga}_{0.5}\text{Ta}_{0.5})\text{O}_3$ solid solutions at 100kHz, showing that the tunability decreases as the dielectric $\text{Sr}(\text{Ga}_{0.5}\text{Ta}_{0.5})\text{O}_3$ content increases. The decrease in the dielectric constant and tunability of $0.9\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-}0.1\text{Sr}(\text{Ga}_{0.5}\text{Ta}_{0.5})\text{O}_3$ results from the Ga and Ta substitution into B-site Ti and Sr substitution into A-site Ba in barium strontium titanate. $0.9\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-}0.1\text{Sr}(\text{Ga}_{0.5}\text{Ta}_{0.5})\text{O}_3$ has a dielectric tunability 16% under 2.63kV/mm versus a dielectric constant $\epsilon=534$. The tunability of $0.7\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-}0.3\text{Sr}(\text{Ga}_{0.5}\text{Ta}_{0.5})\text{O}_3$ drops to be 5.7% under 2.63 kV/mm. The microwave dielectric properties of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-Sr}(\text{Ga}_{0.5}\text{Ta}_{0.5})\text{O}_3$ solid solutions were listed in Table 4. With the increase of $\text{Sr}(\text{Ga}_{0.5}\text{Ta}_{0.5})\text{O}_3$ content, the dielectric constant decrease and the $Q\times f$ value increase. The $Q\times f$ value of the solid solution is not high except $0.5\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-}0.5\text{Sr}(\text{Ga}_{0.5}\text{Ta}_{0.5})\text{O}_3$. The low relative density maybe is the main reason: the relative density of $0.9\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-}0.1\text{Sr}(\text{Ga}_{0.5}\text{Ta}_{0.5})\text{O}_3$, $0.8\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-}0.2\text{Sr}(\text{Ga}_{0.5}\text{Ta}_{0.5})\text{O}_3$ and $0.7\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-}0.3\text{Sr}(\text{Ga}_{0.5}\text{Ta}_{0.5})\text{O}_3$ ceramics sintered at 1600°C for 3h is 82%, 89% and 88%, respectively.

Sr(Ga _{0.5} Ta _{0.5})O ₃ content(mol%)	f ₀ (GHz)	ε	tanδ	Q×f(GHz)
10	1.73	592.4	0.015	115
20	2.05	375.3	0.013	158
30	2.49	236.9	0.012	208
50	4.68	79.6	0.0039	1200

Table 4. Microwave dielectric properties of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-Sr}(\text{Ga}_{0.5}\text{Ta}_{0.5})\text{O}_3$ solid solutions

3.2 $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ solid solution

$\text{La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ with low dielectric constant and loss tangent can form solid solutions with BaTiO_3 or SrTiO_3 in the whole compositional range (Avdeev 2002; Lee 2000). As shown in Fig. 21., $\text{La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ form solid solution with $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ (Xu et al. 2009).

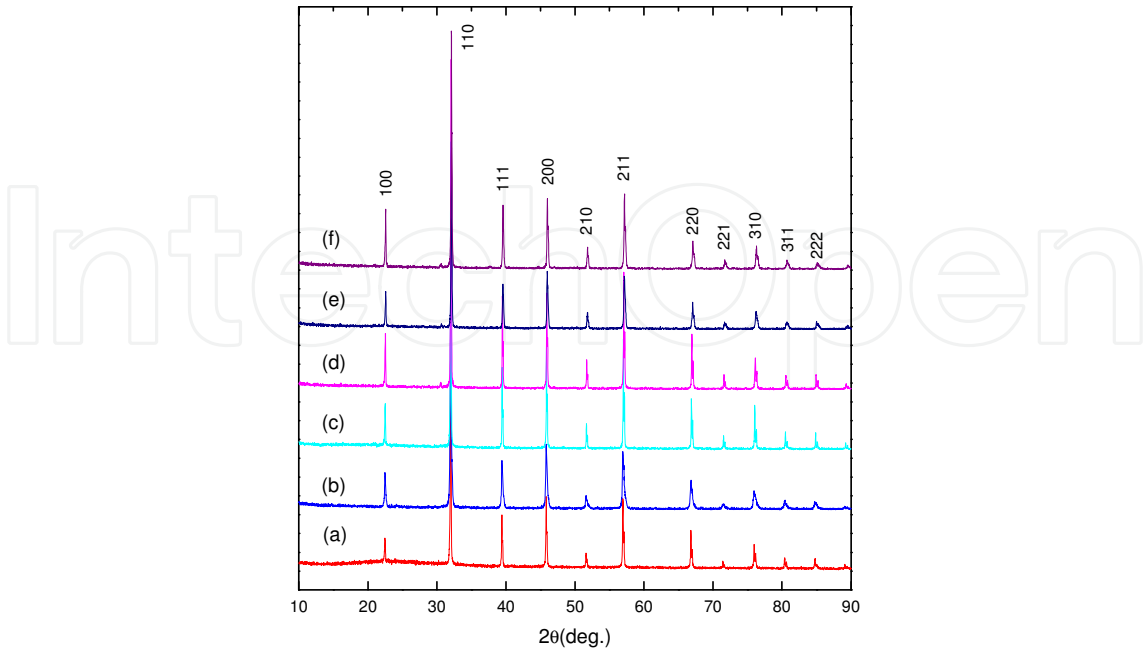


Fig. 21. The XRD patterns of (a) 10, (b) 20, (c) 30, (d) 40, (e) 50 and (f) 60 mol% $\text{La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ mixed $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ ceramics.

The microwave dielectric properties of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-La}(\text{Zn}_{0.5}\text{Ti}_{0.5})\text{O}_3$ ceramics were investigated. For different composition, the optimal sintering temperature is different. If the sintering temperature exceeds the corresponding value, the sample's rim and then interior became dark-blue in color, due to partial reduction of Ti^{4+} (d^0) to Ti^{3+} (d^1) associated with the oxygen loss from the lattice. Fig. 22 show the dielectric constant and Qf of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ ceramics sintered at optimal temperature. ϵ_r decreases with the increase of $\text{La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ content, from $\epsilon_r=338.2$ for $0.9\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-}0.1\text{La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ to $\epsilon_r=47$ for $0.4\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-}0.6\text{La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$. Qf value increases with increasing amounts of $\text{La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$. High Qf value of 9509 GHz with dielectric constant of 46.7 was obtained for $0.4\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-}0.6\text{La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ at 5.69 GHz.

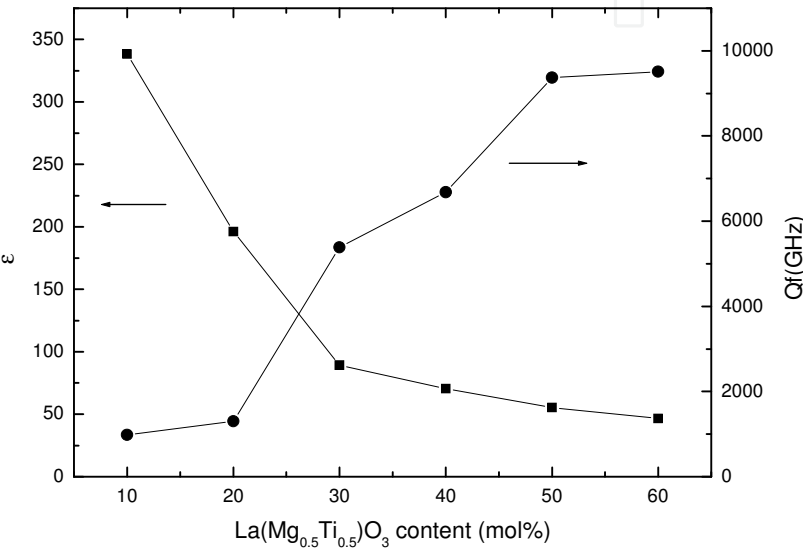


Fig. 22. Dielectric constant and quality factor of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ compositions as a function of $\text{La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ content.

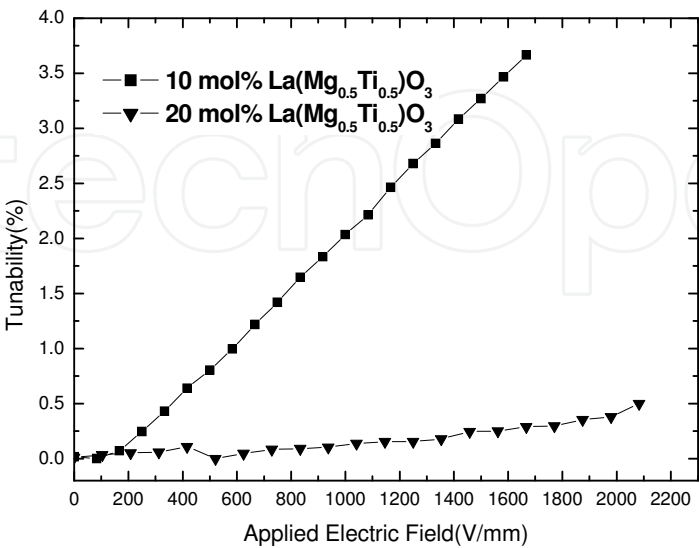


Fig. 23. The tunability of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ compositions measured at 100kHz and room temperature.

The tunability of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ ceramics is shown in Fig. 23. $\text{La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ decreases the tunability of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ abruptly. The tunability of $0.9\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-}0.1\text{La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ is only 3.7% under 1.67 kV/mm, although its Q_f reaches 979GHz. Increasing $\text{La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ content decreases the tunability further: the tunability of $0.8\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-}0.2\text{La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ is 0.5% under 2.08 kV/mm.

The typical FESEM images of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ ceramics sintered at optimal temperature and the energy dispersive spectroscopy of $0.9\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-}0.1\text{La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ were shown in Fig. 24. For $0.4\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-}0.6\text{La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$, dense ceramics were obtained, but higher porosity can be observed for the other three compositions. The high Q_f value of $0.4\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-}0.6\text{La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ can be related to its higher relative density. The chemical composition calculated from energy dispersive spectroscopy were listed in Table 5. We can see that the measured At% is consistent with the theoretical value within the error range. The result also proves the formation of solid solution further.

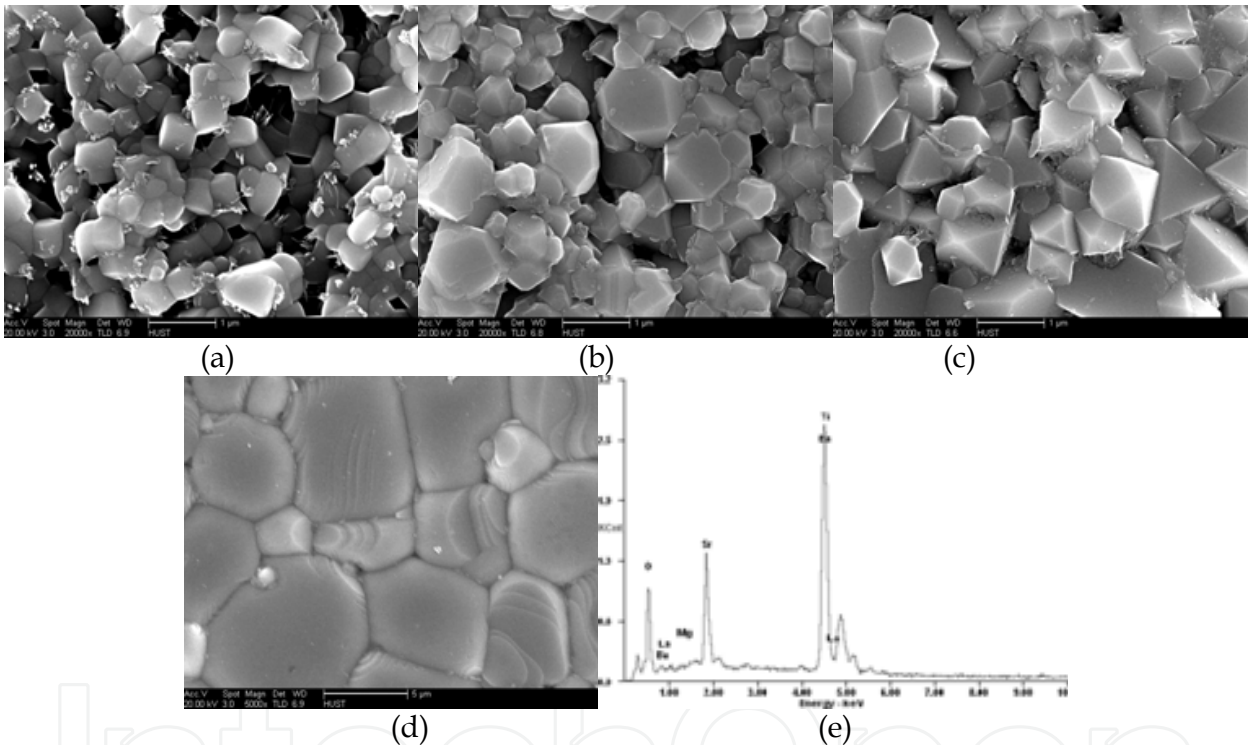


Fig. 24. FESEM images of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ ceramics and the energy dispersive spectroscopy of $0.9\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-}0.1\text{La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ (f). From (a) to (d), $\text{La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ content is 10, 20, 30 and 60 mol%, respectively.

Element	Wt%	At%	Theoretical At%
OK	21.15	56.99	60.61
MgK	00.53	0.95	1.01
SrL	18.65	9.18	7.27
BaL	28.76	9.03	10.91
TiK	24.19	21.77	19.19
LaL	06.71	2.08	2.02

Table 5. The chemical composition of $0.9\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-}0.1\text{La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$

3.3 Ba_{0.6}Sr_{0.4}TiO₃-La(Zn_{0.5}Ti_{0.5})O₃ and Ba_{0.6}Sr_{0.4}TiO₃-Nd(Mg_{0.5}Ti_{0.5})O₃ solid solution
La(Zn_{0.5}Ti_{0.5})O₃ have a comparatively small dielectric constant ($\epsilon=34$ at 10GHz), a negative temperature coefficient of the resonance frequency ($\tau_f=-52\text{ppmK}^{-1}$) and a low dielectric loss

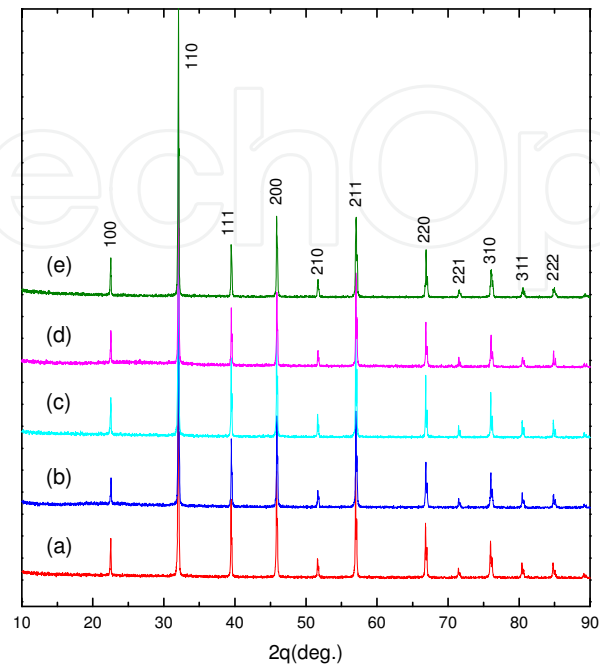


Fig. 25. The XRD patterns of (a) 10, (b) 20, (c) 30, (d) 40, and (e) 50 mol% La(Zn_{0.5}Ti_{0.5})O₃ mixed Ba_{0.6}Sr_{0.4}TiO₃ ceramics.

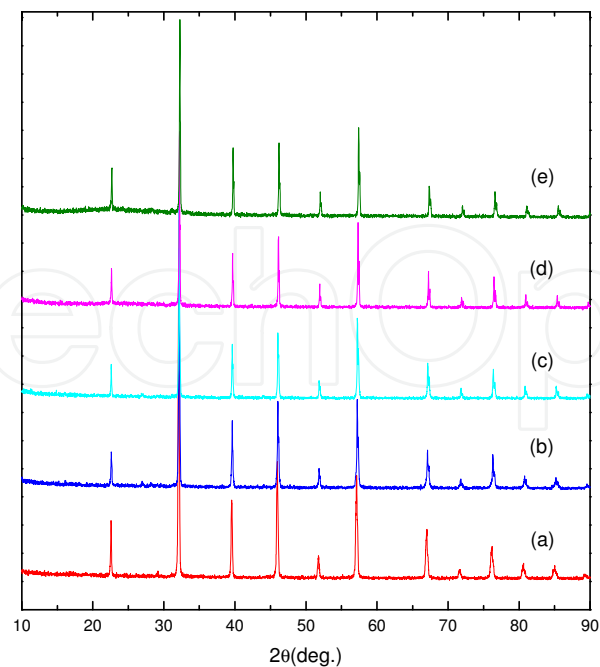


Fig. 26. The XRD patterns of (a) 10, (b) 20, (c) 30, (d) 40, and (e) 50 mol% Nd(Mg_{0.5}Ti_{0.5})O₃ mixed Ba_{0.6}Sr_{0.4}TiO₃ ceramics.

($Q_f=59000\text{GHz}$) (Cho et al. 1997). For $\text{Nd}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$, the corresponding value is 26, -49 ppmK^{-1} and 36900GHz , respectively (Cho et al. 1999). XRD analysis showed that they can form solid solution with $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ (Fig. 25 and 26), but their microwave dielectric properties is inferior to that of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$.

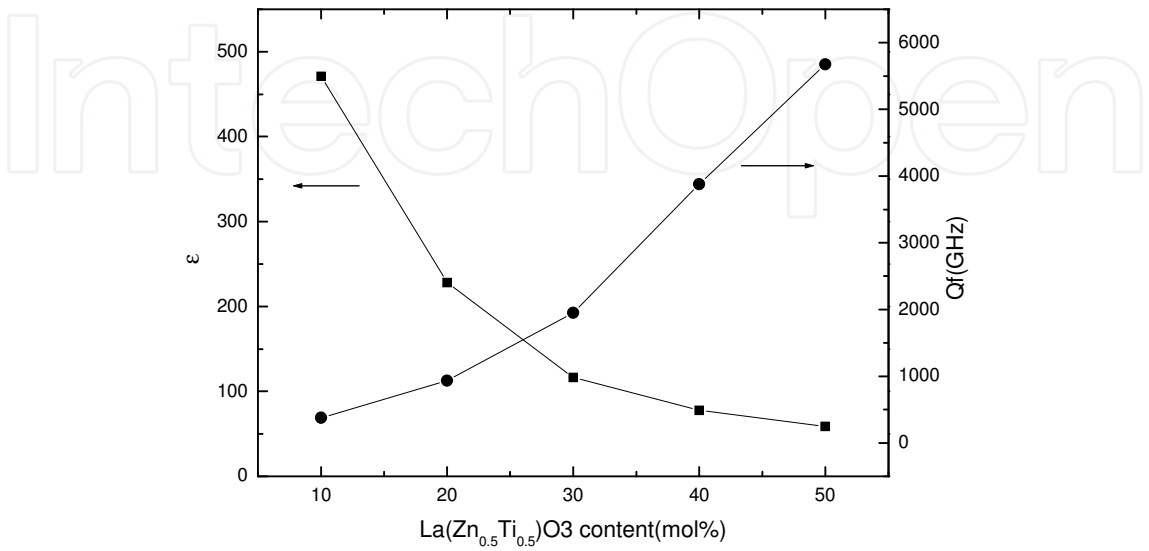


Fig. 27. Dielectric constant and quality factor of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-La}(\text{Zn}_{0.5}\text{Ti}_{0.5})\text{O}_3$ compositions as a function of $\text{La}(\text{Zn}_{0.5}\text{Ti}_{0.5})\text{O}_3$ content.

Fig. 27 show the dielectric constant and Q_f of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-La}(\text{Zn}_{0.5}\text{Ti}_{0.5})\text{O}_3$ ceramics. The dielectric constant of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-La}(\text{Zn}_{0.5}\text{Ti}_{0.5})\text{O}_3$ solid solution decrease as the $\text{La}(\text{Zn}_{0.5}\text{Ti}_{0.5})\text{O}_3$ content increases. The Q_f values of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-La}(\text{Zn}_{0.5}\text{Ti}_{0.5})\text{O}_3$ increase monotonously with increasing $\text{La}(\text{Zn}_{0.5}\text{Ti}_{0.5})\text{O}_3$ content. The highest Q_f value of 5674 GHz was achieved in $0.5\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-}0.5\text{La}(\text{Zn}_{0.5}\text{Ti}_{0.5})\text{O}_3$ but reduced to 377GHz for $0.9\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-}0.1\text{La}(\text{Zn}_{0.5}\text{Ti}_{0.5})\text{O}_3$. The effect of $\text{La}(\text{Zn}_{0.5}\text{Ti}_{0.5})\text{O}_3$ on the microwave dielectric properties of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ solid solution is similar to that of $\text{La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$. The Q_f value of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-La}(\text{Zn}_{0.5}\text{Ti}_{0.5})\text{O}_3$ is lower obviously than that of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ system although their relative density is higher than that of the corresponding $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$.

Nd(Mg _{0.5} Ti _{0.5})O ₃ content (mol%)	sintering temperature (°C)	f ₀ (GHz)	ε	Q _f (GHz)
20	1500	2.68	198.3	535
20	1550	2.83	193.0	615
30	1500	4.05	93.0	880
30	1550	4.30	94.7	1137

Table 6. Microwave dielectric properties of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-Nd}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ solid solutions

Table 6 lists the microwave dielectric properties of some $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-Nd}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ ceramics. Although increasing $\text{Nd}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ content can increase the Q_f value, the Q_f value is not ideal: they are even lower than that of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-La}(\text{Zn}_{0.5}\text{Ti}_{0.5})\text{O}_3$ system.

The tunability of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-La}(\text{Zn}_{0.5}\text{Ti}_{0.5})\text{O}_3$ ceramics is shown in Fig. 28. The tunability of $0.9\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-}0.1\text{La}(\text{Zn}_{0.5}\text{Ti}_{0.5})\text{O}_3$ is only 2.7% under 1.67 kV/mm. It is even smaller than that of $0.9\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-}0.1\text{La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ although $0.9\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-}0.1\text{La}(\text{Zn}_{0.5}\text{Ti}_{0.5})\text{O}_3$ has higher dielectric constant and loss tangent than that of $0.9\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-}0.1\text{La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$. Similarly, increasing $\text{La}(\text{Zn}_{0.5}\text{Ti}_{0.5})\text{O}_3$ content decreases the tunability of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-La}(\text{Zn}_{0.5}\text{Ti}_{0.5})\text{O}_3$ further. We can see that the dielectric properties of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ is better than that of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-La}(\text{Zn}_{0.5}\text{Ti}_{0.5})\text{O}_3$.

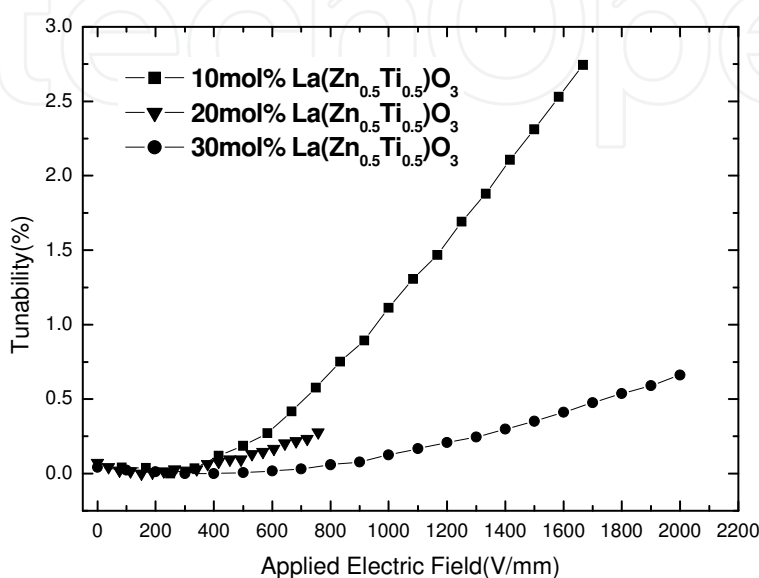


Fig. 28. The tunability of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-La}(\text{Zn}_{0.5}\text{Ti}_{0.5})\text{O}_3$ compositions measured at 100kHz and room temperature.

4. Discussion

Forming ferroelectric-dielectric solid solution and composite both can reduce the dielectric constant of ferroelectrics efficiently, but has different effect on the dielectric properties of ferroelectrics. (1). Forming solid solution can decrease the dielectric constant of ferroelectrics more rapidly when the doping content is nearly the same. The dielectric constant of $0.5\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-}0.5\text{La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ is 55, which is far lower than that of 60 wt% MgO-mixed $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ ($\epsilon=118$) (Chang & Sengupta 2002; Sengupta & Sengupta 1999) although the doping content in 60 wt% MgO-mixed $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ is higher and MgO has lower dielectric constant than $\text{La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$. (2). Forming solid solution can improve the loss tangent of ferroelectrics more effectively. The Qf value of $0.5\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-}0.5\text{La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ and 60 wt% MgO-mixed $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ is 9367GHz and 750GHz (Chang & Sengupta 2002; Sengupta & Sengupta 1999), respectively. Even for loose $0.9\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-}0.1\text{La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ ceramics, its Qf value (979GHz) is much higher than that of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-Mg}_2\text{TiO}_4\text{-MgO}$. In the preparation process of microwave dielectric ceramics, the formation of secondary phase should be prevented. (3). Forming multiphase composite can maintain sufficiently high tunability. $0.5\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-}0.5\text{La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ has lost tunability completely, but the tunability of 60 wt% MgO-mixed $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ at 2kV/mm and 8kV/mm is 10% and 38%, respectively (Chang & Sengupta 2002; Sengupta & Sengupta 1999).

Some authors addressed the dielectric response of ferroelectric-dielectric composites theoretically and various composite models were used to evaluate the dielectric constant, tunability, and loss tangent (Astafiev 2003; Sherman et al. 2006; Tagantsev et al. 2006). As Tagantsev stated (Tagantsev et al. 2006), mixing a tunable ferroelectric with a linear dielectric may modify the electrical properties of the material due to mainly two effects: (i) “doping effect”, –effect of doping of the ferroelectric lattice via the substitution of the ions of the host material and (ii) “composite effect” –effects of redistribution of the electric field in the material due to the precipitation of the non-ferroelectric phase at the grain boundaries or in the bulk of the material. The first effect results primarily in a shift and smearing of the temperature anomaly of the permittivity. The second effect leads to a redistribution of the electric field in the material. For ferroelectric-dielectric solid solutions, “composite effect” can be excluded. We can deduce that the addition of low loss perovskite dielectric influenced the chemistry and microstructure of the material, which resulted in the change of dielectric properties of materials. In ferroelectric-dielectric solid solution, a high degree of structural disorder due to random cation arrangement in both A- and B-sites is present, addition of perovskite dielectrics apparently destroys the ferroelectric state, leading to the sharp decrease of tunability. For ferroelectric-dielectric composites, “doping effect” can be ignored. The effect of the dilution-driven field redistribution in the material is the main manifestation of addition of the dielectric into ferroelectrics in two-phase or multiphase composite. The reduction of the volume of ferroelectric, which is responsible for tuning, causes suppression of the tunability of the material. In ferroelectric-dielectric composites, ferroelectrics host lattice remains unchanged and the decrease of tunability is mainly due to ferroelectric dilution. “Destruction” in solid solution and “dilution” in composite has different effect on the tunability. Therefore, forming ferroelectric-dielectric solid solution can cause the decrease of tunability more sharply.

In ferroelectric-dielectric composite, the big contrast in the values of dielectric constants of linear dielectrics and the ferroelectric affects the redistribution of the electric field around the dielectrics. The dielectric constant of the ferroelectric under applied electric field becomes in-homogeneously distributed over the volume of the ferroelectric. The overall tunability of the composite, thus changes. Two competitive phenomena affect the tunable properties of the ferroelectric when it is diluted with a dielectric (Sherman et al. 2006). First, the reduction of the volume of ferroelectric, which is responsible for tuning, will cause suppression of the tunability of the material. Second, the redistribution of the electric field surrounding the linear dielectrics will affect the local tuning of the ferroelectric. Depending on the shape of the linear dielectrics and on the dielectric constants of the components, the impact of each of these two effects on the composite tunability is different and the second effect may be stronger (Sherman et al. 2006). In ferroelectric-dielectric composite $\text{BaZr}_{0.2}\text{Ti}_{0.8}\text{O}_3\text{-Mg}_2\text{SiO}_4\text{-MgO}$, with the increase of Mg_2SiO_4 content and the decrease of MgO content, the volume of ferroelectric $\text{BaZr}_{0.2}\text{Ti}_{0.8}\text{O}_3$ decrease due to smaller density of Mg_2SiO_4 than that of MgO , the tunability of composite will be suppressed. It is the fact as MgO content decreases from 48 wt% to 30 wt%. The anomalous increased tunability in $\text{BaZr}_{0.2}\text{Ti}_{0.8}\text{O}_3\text{-Mg}_2\text{SiO}_4\text{-MgO}$ composite with MgO content < 30wt% can be attributed to redistribution of the electric field. Mg_2SiO_4 and MgO have different dielectric constants, they will have different effects on the redistribution of the electric field. The combination of linear dielectrics with different dielectric constants can result in the change of dielectric constant and loss tangent and even increase the tunability by affecting the redistribution of the electric field in the composite. As MgO content decreases from 30 wt% to 12 wt%, the increase of the

tunability due to redistribution of the electric field exceeds the decrease of the tunability due to ferroelectric dilution, so the tunability of composite ceramics increases anomalously. The almost unchanged tunability in $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-Mg}_2\text{SiO}_4\text{-MgO}$ composite can also be explained. No similar result was observed in $\text{BST-Mg}_2\text{TiO}_4\text{-MgO}$ composite: with the increase of MgO content and the decrease of Mg_2TiO_4 content, the dielectric constant and tunability decrease monotonously. The tunability of $40\text{Ba}_{0.5}\text{Sr}_{0.5}\text{TiO}_3\text{-}48\text{Mg}_2\text{TiO}_4\text{-}12\text{MgO}$ ceramics at 2kV/mm is 16.6%, but the corresponding tunability of $40\text{Ba}_{0.5}\text{Sr}_{0.5}\text{TiO}_3\text{-}12\text{Mg}_2\text{TiO}_4\text{-}48\text{MgO}$ is only 5.7%.

We can also see that ternary compositions ferroelectric-dielectric composite shows some advantages over binary compositions. (1). We can decrease the dielectric constant of ternary composites and remain the tunability almost unchanged (in $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-Mg}_2\text{SiO}_4\text{-MgO}$), even increase the tunability (in $\text{BaZr}_{0.2}\text{Ti}_{0.8}\text{O}_3\text{-Mg}_2\text{SiO}_4\text{-MgO}$), without increasing the content of linear dielectrics. In order to decrease the dielectric constant of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-MgO}$, it is necessary to increase the content of linear dielectrics MgO, and the tunability will decrease inevitably. The tunability of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-MgO}$ decreases with the increase of MgO content (Chang & Sengupta 2002). For $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-Mg}_2\text{SiO}_4\text{-MgO}$ composite, their dielectric constant decrease to 85-97, the tunability at 2kV/mm can be kept at around 10%. (2). The sintering temperature of ternary compositions $\text{BaZr}_{0.2}\text{Ti}_{0.8}\text{O}_3\text{-Mg}_2\text{SiO}_4\text{-MgO}$ and $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-Mg}_2\text{SiO}_4\text{-MgO}$ can be reduced to 1350°C, which is 100°C and 150°C lower than the normal sintering temperature of BST-MgO (~1450°C) and BZT-MgO (~1500°C), respectively. The sintering temperature of $\text{BST-Mg}_2\text{TiO}_4\text{-MgO}$ composite is also lower than that of BST-MgO .

On the other hand, ternary composition is helpful for the formation of ferroelectric-dielectric composite and can prevent from the formation of undesired phase. Forming ferroelectric-dielectric composite is an effective method to reduce the dielectric constant and loss tangent of ferroelectric and maintain higher tunability. The key is that the linear dielectrics with low dielectric constant and loss tangent should not react with ferroelectrics. For binary composition $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-Mg}_2\text{SiO}_4$, it is expected to form ferroelectric ($\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$)-dielectric (Mg_2SiO_4) composite, but undesired impurity phase $\text{Ba}_2(\text{TiO})(\text{Si}_2\text{O}_7)$ is formed among $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-Mg}_2\text{SiO}_4$ composite. $\text{Ba}_2(\text{TiO})(\text{Si}_2\text{O}_7)$ deteriorate the properties of composites. MgO and Mg_2SiO_4 combination can prevent from the formation of $\text{Ba}_2(\text{TiO})(\text{Si}_2\text{O}_7)$ and ferroelectric ($\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$) and dielectric (Mg_2SiO_4 and MgO) composite is obtained. Similarly, Mg_2TiO_4 can react with $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$ ($x=0.5$ and 0.6) to form $\text{BaMg}_6\text{Ti}_6\text{O}_{19}$, but no $\text{BaMg}_6\text{Ti}_6\text{O}_{19}$ formed in $\text{BST-Mg}_2\text{TiO}_4\text{-MgO}$ composite. Therefore, maybe ternary compositions can open a new route to decrease the dielectric constant and loss tangent of ferroelectrics and remain higher tunability. In future work, it is necessary to search new combination of linear dielectrics. Even if one linear dielectric may react with ferroelectrics, some linear dielectrics combination is possible to form ferroelectric-dielectric composite with ferroelectrics. This will expand the select range of linear dielectrics.

The multiple-phase ferroelectric-dielectric composites are useful for tunable microwave applications requiring low dielectric constant and make the impedance match more easily. The ferroelectric-dielectric composite bulk ceramics show promising application, especially in accelerator, as active elements of electrically controlled switches and phase shifters in pulse compressors or power distribution circuits of future linear colliders as well as tuning layers for the dielectric based accelerating structures (Kanareykin et al. 2006, 2009a, 2009b). The ferroelectric bulk ceramics can also be used in ferroelectric lens (Rao et al. 1999).

The ferroelectric-dielectric composites might complicate method to effectively deposit films. Therefore, the advantage of ferroelectric-dielectric solid solution over composite is that single phase materials is favorable for the thin film deposition. At the meantime, the tunability of solid solution can be increase to relatively high level by increasing applied electric field. Generally, linear dielectric with perovskite structure can form solid solution with ferroelectric BST. Different linear dielectrics has different effects on the dielectric properties. Among studied solid solution, $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ shows better properties. It is necessary to increase the density of the solid solution, meanwhile, prevent the reduction of Ti^{4+} .

5. Conclusion

Forming ferroelectric-dielectric composite and solid solution can reduce the dielectric constant of ferroelectrics efficiently, but the mechanisms affecting dielectric properties differ in composites and solid solutions. Forming ferroelectric-dielectric solid solution can improve the loss tangent of ferroelectrics more effectively and is beneficial to film deposition. Forming ferroelectric-dielectric composite is more efficient to decrease the dielectric constant of ferroelectrics to a low value and maintain tunability at a sufficiently high level.

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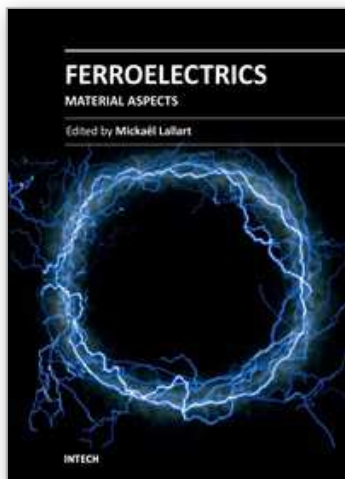
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Ferroelectric materials have been and still are widely used in many applications, that have moved from sonar towards breakthrough technologies such as memories or optical devices. This book is a part of a four volume collection (covering material aspects, physical effects, characterization and modeling, and applications) and focuses on ways to obtain high-quality materials exhibiting large ferroelectric activity. The book covers the aspect of material synthesis and growth, doping and composites, lead-free devices, and thin film synthesis. The aim of this book is to provide an up-to-date review of recent scientific findings and recent advances in the field of ferroelectric materials, allowing a deep understanding of the material aspects of ferroelectricity.

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