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Microwave Dielectric Properties of $ZnNb_2O_6$-$SrTiO_3$ Stacked Resonators

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Microwave dielectric properties of $ZnNb_2O_6$-$SrTiO_3$ (ZN/ST) stacked resonators were investigated. ZN/ST stacked resonators with a zero τ_f have been obtained by adjusting the volume percentage of ST. Resonant frequency decreased from 9.27 GHz for 8.57% ST to 4.94 GHz for 27.27% ST. ε_r increased from 24.6 for 8.57% ST to 66.2 for 27.27% ST. $Q \times f$ rapidly decreased from 48,230 GHz for 8.57% ST to 7798 GHz for 15.79% ST and then 962 GHz for 27.27% ST. A temperature-stable ZN/ST stacked resonator with τ_f = 0 ppm/°C was obtained for 8.57% ST. τ_f increased to 678 ppm/°C for 27.27% ST. Dielectric properties: ε_r = 24.6, $Q \times f$ = 48,230 GHz and τ_f = 0 ppm/°C are obtained for a ZN/ST stacked resonator with 8.57% ST.

Key words: Microwave dielectric ceramics, layered structure, $ZnNb_2O_6$, $SrTiO_3$

INTRODUCTION

Many microwave dielectric ceramics with excellent quality factors ($Q \times f$) and dielectric constants (ε_r) had been developed.[1] However, the poor temperature stability has limited the practical application for some of these ceramics. The temperature coefficient of the resonant frequency (τ_f) of these ceramics needs to be tuned. Mixing two materials with opposite τ_f to form a solid solution had been the frequently used tuning method. Unfortunately, not all microwave dielectric ceramics can be improved by this method. Problems such as possible incompatibility of ionic radius, ionic charge, or crystal structure will result in some undesired secondary phases and degrade the microwave dielectric properties.[2–4]

Stacked resonators used at microwave frequencies have been widely investigated after the report of obtaining temperature compensation by stacking two cylindrical resonators by Tsironis and Panker[5] in 1983. A stacked resonator made of two different materials with τ_f of opposite signs is possible to be temperature-stable. Breeze et al.[6] reported layered Al_2O_3-TiO_2 composite dielectric resonators. The application of a film of TiO_2 which has a τ_f of +450 ppm/°C produces an Al_2O_3-TiO_2 composite in which the τ_f can be tuned to be zero over a wide temperature range. Sebastian et al.[2] studied dielectric resonators (DRs) with positive τ_f stacked with resonators with negative τ_f. The experiment is performed with varying volume fractions of $Ba_5Nb_4O_{15}$ as the positive τ_f DR and $Sr(Y_{1/2}Nb_{1/2})O_3$ and $5ZnO$-$2Nb_2O_5$ as the negative τ_f DR materials. They found τ_f of the stacked resonator can be tuned to 0 or to a desired value by adjusting the volume fraction of the positive and negative τ_f materials. Chen et al.[7] reported a near-zero τ_f can be achieved in the layered complex dielectric structures of $MgTiO_3/CaTiO_3$ with low dielectric loss and greater dielectric constant than that in $MgTiO_3/CaTiO_3$ solid solution. Li et al.[8] studied $Ca(Mg_{1/3}Nb_{2/3})O_3/Ba(Zn_{1/3}Nb_{2/3})O_3$ layered dielectric resonators. They found a good combination of microwave dielectric characteristics with an ε_r of 34.33–34.52, a $Q \times f$ value of 58,800–62,080 GHz, and a near-zero τ_f could be achieved by adjusting the volume fraction of $Ba(Zn_{1/3}Nb_{2/3})O_3$. They also studied $MgTiO_3/SrTiO_3$ ($MgTiO_3$/ST) layered ceramics with a 25–75% volume percentage of ST

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and found that with increasing ST thickness fraction, the resonant frequency decreased, while ε_r and τ_f increased for the bi-layer ceramics.[9] Kuo[10] studied $MgTiO_3$/ST layered ceramics with 8.57–27.27% volume percentages of ST and found excellent dielectric properties: $\varepsilon_r = 18.9$, $Q \times f = 58145$ GHz and $\tau_f = 8.27$ ppm/°C for 8.57% ST. Zhou et al.[11] reported piling up and cofiring layered complex structures of $Bi_2(Zn_{2/3}Nb_{4/3})O_7$ and $BiNbO_4$ ceramics. The piled up complex showed better properties than the cofired $Bi_2(Zn_{2/3}Nb_{4/3})O_7/BiNbO_4$ ceramics.

$ZnNb_2O_6$ (ZN) ceramics were reported to exhibit excellent dielectric properties: $\varepsilon_r = 25$, $Q \times f = 83700$ GHz and $\tau_f = -56.1$ ppm/°C.[12] ST ceramics exhibit $\varepsilon_r \sim 205$, $Q \times f \sim 4200$ GHz and $\tau_f \sim 1700$ ppm/°C.[13] ST is generally introduced into microwave dielectric ceramics with a negative τ_f to obtain near-zero τ_f. In our previous study, dense ZN ceramics of 5.55 g/cm^3 (98.7% of theoretical value) and dielectric properties: $\varepsilon_r = 23.5$, $Q \times f = 61916$ GHz and $\tau_f \sim -50$ ppm/°C were obtained after 1200°C/2 h sintering via a reaction-sintering process.[14,15] To the best of our knowledge, there is no report about the temperature-stable ZN-ST microwave dielectric ceramics prepared by forming a solid solution or stacked resonators. In this study, we try to obtain ZN-ST stacked resonators with a near-zero τ_f by adjusting the volume fraction of ST.

EXPERIMENTAL PROCEDURES

ZN and ST ceramics used in this study were prepared via reaction-sintering process individually. The preparation characterization and properties are reported elsewhere.[14–16] 1200°C/2-h-sintered ZN (5.55 g/cm^3, 98.7% of theoretical density) and 1350°C/2-h sintered ST (4.44 g/cm^3, 86.8% of theoretical density) were used. ZN and ST pellets with nearly the same diameter of 9.65 mm were thinned to different thickness as desired, and polished well with paralleling and smooth surfaces. 4-mm-thick ZN and 1.5-mm-thick ST were stacked together using the UHU hart kunststoff glue (temperature-resistant from −30°C up to +90°C, Germany) ~0.02 mm thick as shown in Fig. 1.

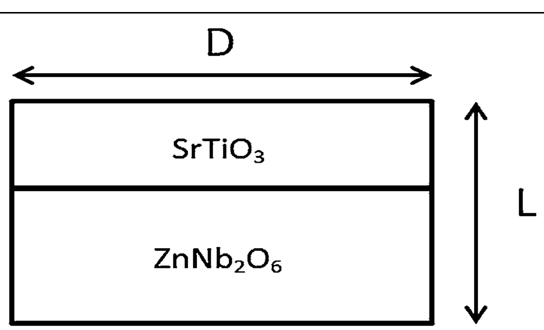

Fig. 1. Configuration of ZN/ST stacked resonator. *D* diameter; *L* total thickness.

Dielectric properties at microwave frequencies were measured using the Hakki–Coleman dielectric resonator method.[17] Dielectric resonators were positioned between two conducting brass plates with one plate being adjustable. The dielectric constant can be calculated from dimensions of the resonator and the accurately measured resonant frequency of the TE_{011} mode. An Agilent 8720ES network analyzer was used to measure the frequencies. τ_f at microwave frequency was measured in the temperature range from 25°C to 80°C, and calculated using Eq. 1.

$$\tau_f = (f_{80} - f_{25})/(f_{25} \times 55) \times 10^6 (\text{ppm}/°\text{C}) \qquad (1)$$

where f_{80} and f_{25} are the $TE_{01\delta}$ resonant frequencies at 80°C and 25°C, respectively. The volume percentage of ST was altered by thinning the thickness of the ST layer in the same ZN/ST stacked resonator after the measurement of ε_r, $Q \times f$ and τ_f. The thickness and the corresponding volume fraction are listed in Table I.

RESULTS AND DISCUSSION

The resonant frequency f_{25} for ZN/ST stacked resonators at various volume percentages of ST is shown in Fig. 2. f_{25} decreased from 9.27 GHz for 8.57% ST to 4.94 GHz for 27.27% ST. f_{25} decreased ~0.232 GHz on average for 1% of ST increase. A microwave dielectric ceramic with a higher ε_r usually has a lower resonant frequency. As ε_r of ST is much higher than ε_r of ZN, f_{25} decreased with an increased volume percentage of ST for the ZN/ST stacked resonators in Fig. 2. Referring to Ref. 2, the resonant frequency of a $Ba_5Nb_4O_{15}$ resonator increased from 4.9275 GHz for 5.69 mm in length and 9.64 mm in diameter to 5.1804 GHz for 4.49 mm in length and 9.64 mm in diameter. It increased about 5.13% when the diameter/length ratio was varied from 1.69 to 2.15. On the other hand, the resonant frequency of a $5ZnO\text{-}2Nb_2O_5$ resonator increased from 6.475 GHz for 5.71 mm in length and 9.67 mm in diameter to 6.738 GHz for 4.77 mm in length and 9.67 mm in diameter. It increased about 4.06% when the diameter/length ratio was varied from 1.69 to 2.03. When the diameter/length ratio varied from 1.75 to 2.20 (5.5 mm to 4.375 mm in total length), we thought the resonant frequency of ZN/ST stacked resonator with major part of ZN would increase less than 4% considering the similar dielectric constant for the $5ZnO\text{-}2Nb_2O_5$ resonator (22) and ZN resonator (23.5). However, the resonant frequency of the ZN/ST stacked resonator increased 87.65% when the diameter/length ratio was varied from 1.75 to 2.20, as per results in Table II. This indicates that the decreased ST thickness seems to be the major affecting factor rather than the increased diameter/length ratio. The resonant frequency at 20°C decreased from ~4.85 GHz for 25% ST to ~4.1 GHz for 33.3% ST (~0.09 GHz on average for 1% ST

Table I. Thickness and volume percentage (vol.%) of ST for ZN/ST stacked resonators

$ZnNb_2O_6/SrTiO_3$ (mm)	4/1.5	4/1.125	4/0.75	4/0.375
$SrTiO_3$ vol.%	27.27%	21.95%	15.79%	8.57%

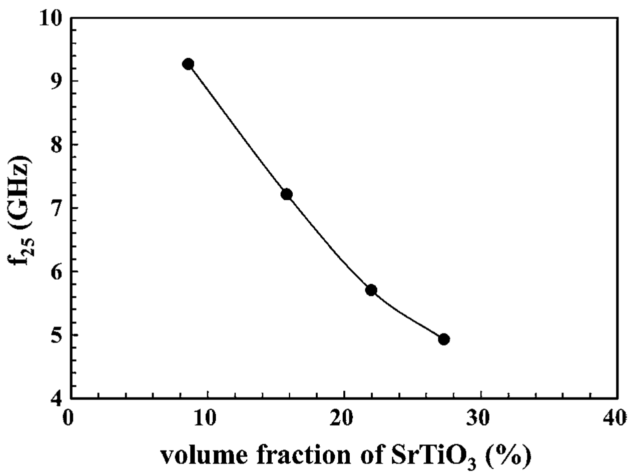

Fig. 2. Resonant frequency at 25 °C for ZN/ST stacked resonator for various volume percentage of $SrTiO_3$.

increase) for $MgTiO_3$/ST layered ceramics reported by Li et al.[9] This is smaller than ~0.146 GHz on average for 1% ST increase from 21.95% to 27.27% ST in Fig. 2. the resonant frequency at 25°C decreased from 10.54 GHz for 8.57% ST to 5.35 GHz for 27.27% ST (~0.277 GHz on average for 1% ST increase) for $MgTiO_3$/ST layered ceramics reported by Kuo.[10] The decreasing tendency of resonant frequency for $MgTiO_3$/ST layered ceramics by cofiring is different from $MgTiO_3$/ST stacked by glue. The resonant frequency decreases in a similar fashion when ST is stacked with ZN and $MgTiO_3$ by glue.

Figure 3 shows ε_r of ZN/ST stacked resonators for various volume fractions of ST. ε_r increased from 24.6 for 8.57% ST to 66.2 for 27.27% ST. ε_r increased ~2.22 on average for 1% ST increase. ε_r increased from ~70.8 for 25% ST to ~99.6 for 33.3% ST (~3.47 on average for 1% ST increase) for $MgTiO_3$/ST layered ceramics reported by Li et al.[9] ST with a lower density (4.44 g/cm^3, 86.8% of theoretical value) used in this study may be the reason why the increasing tendency of ε_r for 1% ST increase is lower than ~3.47 reported by Li et al.[9] Referring to dielectric properties: $\varepsilon_r \sim 310$, $Q \times f \sim 5700$ GHz and $\tau_f \sim 1660$ ppm/°C for ST ceramics reported by Li et al.,[9] ε_r in Fig. 3 is estimated to increase ~3.4 on average for 1% ST increase between 27.27% and 100%. This is close to ~3.47 on average for 1% ST increase between 25% and 33.3% reported by Li et al.[9] ε_r increased from 18.9 for 8.57% ST to 56 for 27.27% ST (1.98 on average for 1% ST increase) for $MgTiO_3$/ST layered ceramics reported by Kuo.[10] ε_r increases in a similar tendency when ST is stacked with ZN and $MgTiO_3$ by glue.

$Q \times f$ for ZN/ST stacked resonators for various volume percentages of ST is shown in Fig. 4. $Q \times f$ decreased in a different tendency for lower (< 15.79%) and higher (> 15.79%) volume percentages of ST. $Q \times f$ rapidly decreased from 48,230 GHz for 8.57% ST to 7798 GHz for 15.79% ST and then 962 GHz for 27.27% ST. $Q \times f$ decreased ~2530 GHz on average for 1% ST increase between 8.57% and 27.27% ST. ST with a lower density (86.8% of theoretical value) and a lower $Q \times f$ for 0% ST (61916 GHz for ZN; ~92000 GHz for $MgTiO_3$ in Ref. 9) used in this study may be the reason why the decreasing tendency of $Q \times f \sim 2530$ GHz for 1% ST increase is lower than ~3565 GHz reported by Li et al.[9] It is noted that $Q \times f$ for ZN/ST stacked resonators with 21.95% and 27.27% ST are less than $Q \times f \sim 5700$ GHz of ST reported by Li et al.[9] Li et al.[9] observed $Q \times f$ decreased from ~92,000 GHz at 0% ST to ~2890 GHz at 25% ST (~3565 GHz on average for 1% ST increase) then increased to ~3760 GHz at 33.3% ST for $MgTiO_3$/ST layered ceramics. They found $Q \times f$ of the layered ceramics is significantly lower than those of $MgTiO_3$ and ST and is not a combination of those of $MgTiO_3$ and ST. They thought the different thermal expansion coefficients of $MgTiO_3$ and ST cause the residual stresses in the $MgTiO_3$/ST interface, and the residual stresses shall be responsible for the different behavior of the $Q \times f$ value. We thought the residual stress is also the reason why $Q \times f$ for ZN/ST stacked resonators with 21.95% and 27.27% ST are less than $Q \times f$ of ST in this study. Kuo[10] found $Q \times f$ decreased from 58,145 GHz for 8.57% ST to 898 GHz for 27.27% ST (3061 GHz on average for 1% ST increase) for $MgTiO_3$/ST layered ceramics. A lower $Q \times f$ for 0% ST (61,916 GHz for ZN) used in this study may be the reason why the decreasing tendency of $Q \times f \sim 2530$ GHz for 1% ST increase is lower than 3061 GHz reported by Kuo (92,000 GHz for $MgTiO_3$).[10]

τ_f values for ZN/ST stacked resonators of various volume percentages of ST are shown in Fig. 5. τ_f increased in a different tendency for lower (< 15.79%) and higher (> 15.79%) volume percentage of ST. A temperature-stable ZN/ST stacked resonator with $\tau_f = 0$ ppm/°C is obtained for 8.57% ST. τ_f increased to 678 ppm/°C for 27.27% ST (~36.25 ppm/°C on average for 1% ST increase). Li et al.[9] found τ_f increased from ~1010 ppm/°C for 25% ST to ~1039 ppm/°C for 33.3% ST (~3.03 ppm/

Table II. Microwave dielectric properties of ZN/ST stacked resonators

$SrTiO_3$ vol.%	Resonant frequency at 25°C (GHz)	ε_r	$Q \times f$ (GHz)	τ_f (ppm/°C)
27.27%	4.94	66.2	962	678
21.95%	5.71	54	1654	486
15.79%	7.21	36.7	7798	157
8.57%	9.27	24.6	48,230	0

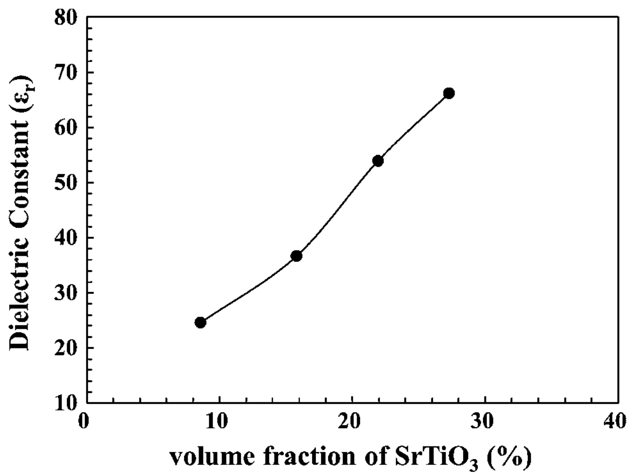

Fig. 3. ε_r of ZN/ST stacked resonator for various volume percentage of $SrTiO_3$.

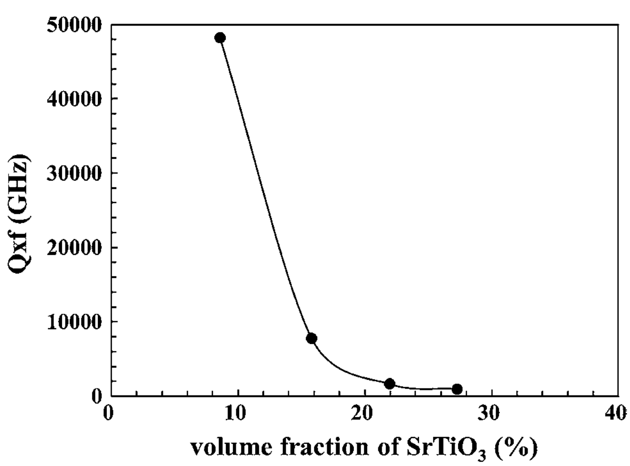

Fig. 4. $Q \times f$ for ZN/ST stacked resonator for various volume percentage of $SrTiO_3$.

°C on average for 1% ST increase) for $MgTiO_3$/ST layered ceramics. Referring to dielectric properties: $\varepsilon_r \sim 310$, $Q \times f \sim 5700$ GHz and $\tau_f \sim 1660$ ppm/°C for ST ceramics reported by Li et al.,[9] τ_f in Fig. 5 is estimated to increase ~13.5 ppm/°C on average for 1% ST increase between 27.27% and 100% ST. This is still higher than ~8.61 ppm/°C on average for 1% ST increase between 25% and 100% ST reported by Li et al.[9] ST affected τ_f in a different tendency for ZN/ST stacked by glue and $MgTiO_3$/ST stacked by cofiring. Kuo found τ_f increased from 8 ppm/°C for

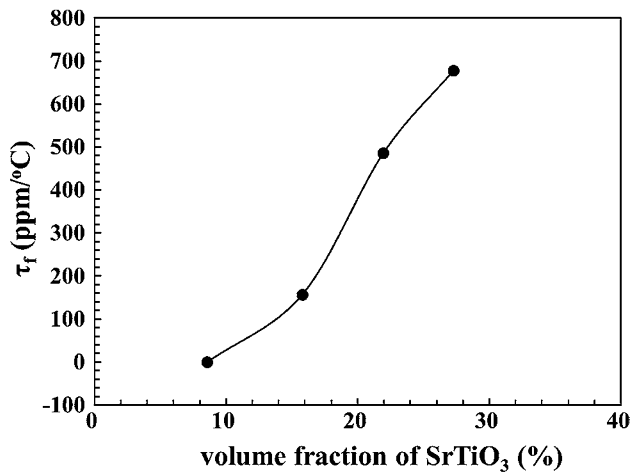

Fig. 5. τ_f for ZN/ST stacked resonator for various volume percentage of $SrTiO_3$.

8.57% ST to 843 ppm/°C for 27.27% ST (~44.65 ppm/°C on average for 1% ST increase) for $MgTiO_3$/ST stacked by glue.[10] τ_f increases in a similar tendency when ST is stacked with ZN and $MgTiO_3$ by glue.

The dielectric properties for microwave dielectric ceramics containing two phases were suggested to obey the well-known mixing rules.[18]

$$\ln \varepsilon_r = v_1 \ln \varepsilon_1 + v_2 \ln \varepsilon_2 \quad (2)$$

$$Q^{-1} = v_1 Q_1^{-1} + v_2 Q_2^{-1} \quad (3)$$

$$\tau_f = v_1 \tau_{f1} + v_2 \tau_{f2} \quad (4)$$

where v_1 and v_2 represent the volume fraction of phase 1 and phase 2 in microwave dielectric ceramics, respectively. As the thickness of the glue and the volume percentage of glue (0.362–0.455%) are much lower than ZN (72.46–91.01%) and ST (8.53–27.17%), it is reasonable to neglect the effect of the glue on the effective value of the dielectric constant of the stacked material according to the mixing rules ($\ln \varepsilon_r = v_1 \ln \varepsilon_1 + v_2 \ln \varepsilon_2 + v_3 \ln \varepsilon_3$). Microwave dielectric properties near 0 ppm/°C of ZN/ST stacked resonators and calculated according the mixing rule are listed in Table III. According to the mixing rule, the composition for ZN/ST by forming a solid solution should be 0.951ZN-0.049ST. Microwave dielectric properties estimated are $\varepsilon_r = 25.3$,

Table III. Microwave dielectric properties near 0 ppm/°C of ZN/ST and $MgTiO_3$/ST stacked resonators and calculated according the mixing rule

Resonators	ε_r	$Q \times f$ (GHz)	τ_f (ppm/°C)	ST vol.%	References
ZN/ST stack	24.6	48,230	0	8.57	This study
ZN/ST mixing rule 0.951ZN-0.049ST	25.3	48,060	0	2.924 (4.926 mol%)	18
$MgTiO_3$/ST stack	18.9	58,145	8.27	8.57	10
$MgTiO_3$/ST mixing rule 0.975$MgTiO_3$-0.025ST	21	63,770	0	2.924 (2.524 mol%)	18
0.964$MgTiO_3$-0.036ST Solution	20.76	71,000	−1.27	4.164	18
ZN	23.5	61,916	−50	0	14
$MgTiO_3$[a]	~19.4	~92,000	~−50	0	9
ST[a]	~310	~5700	~1660	100	9

[a]Estimated by a linear interpolation method referring to plots in Ref. 9.

$Q \times f$ = 48060 GHz and τ_f = 0 ppm/°C. These are very close to properties of ZN/ST stacked resonators obtained in this study. For ZN/ST stacked resonators, a volume percentage of ST 8.57% is needed. This is higher than the volume percentage of ST 2.924% needed for ZN/ST resonators by forming a solid solution. ST with a lower density (4.44 g/cm^3, 86.8% of theoretical value) used in this study may be one reason for the higher ST volume percentage. In using the mixing rule for calculating microwave dielectric properties, full dense ZN and ST ceramics are considered. Therefore, more ST volume is necessary in this study than the needed ST volume calculated according the mixing rule. When a full dense ST is considered for ZN/ST stacked resonators in this study, 7.44% ST is needed. This is still much higher than 2.924% ST for ZN/ST resonators by forming a solid solution. This implies the mechanisms for tuning τ_f in ZN/ST resonators via a stack structure and via a solid solution are different. Microwave dielectric properties near 0 ppm/°C of $MgTiO_3$/ST resonators via a stack structure, a solid solution and calculated according the mixing rule are also listed in Table III for comparison. According to the mixing rule, the composition for $MgTiO_3$/ST by forming a solid solution should be 0.975$MgTiO_3$-0.025ST (2.924% volume percentage of ST). Microwave dielectric properties estimated are ε_r = 21, $Q \times f$ = 63,770 GHz and τ_f = 0 ppm/°C. Cho et al.[19] reported 0.964$MgTiO_3$-0.036ST (4.164% volume percentage of ST) with properties close to results calculated according the mixing rule. However, 4.164% volume percentage of ST is much higher than 2.924% calculated according the mixing rule. $MgTi_2O_5$ phase formed in 0.964$MgTiO_3$-0.036ST prepared by Cho et al.[19] would decrease the content of ST formation. Therefore, more ST is needed to reach a near 0 ppm/°C τ_f. Li et al.[9] reported $\varepsilon_r \sim 70.8$, $Q \times f \sim 2890$ GHz and $\tau_f \sim 1010$ ppm/°C for $MgTiO_3$/ST layered ceramics with 25% volume percentage of ST. Kuo obtained excellent dielectric properties: ε_r = 18.9, $Q \times f$ = 58145 GHz and τ_f = 8.27 ppm/°C for $MgTiO_3$/ST stacked by glue (8.57% volume percentage of ST).[10] Similar to ZN/ST stacked by glue, a higher volume percentage of ST (8.57%) is needed in $MgTiO_3$/ST stacked by glue than in $MgTiO_3$/ST resonators by forming a solid solution (2.924%). This implies the mechanisms for tuning τ_f in $MgTiO_3$/ST resonators via a stack structure and via a solid solution are also different.

Referring to Ref. 3, $MgTiO_3$/$CaTiO_3$ and $CaTiO_3$/$MgTiO_3$ stacks with 25–75% (volume percentage) $CaTiO_3$ showed the same resonant frequency, ε_r, and τ_f while $Q \times f$ did not alter obviously. Referring to Ref. 2, the DR material in the bottom of the stack has greater influence on the resultant τ_f, although ε_r and $Q \times f$ are not affected for $Ba_5Nb_4O_{15}$/5ZnO-2Nb_2O_5 and 5ZnO-2Nb_2O_5/$Ba_5Nb_4O_{15}$ stacks. τ_f is more positive when $Ba_5Nb_4O_{15}$ (τ_f = 78 ppm/°C) was mounted at the bottom. We thought the temperature of the DR material in the bottom increased faster (heat transfers faster via contact than via radiation) than the DR material in the top because the bottom DR is sandwiched by the top DR and the sample support (such as sapphire). When DR with a positive τ_f was in the bottom, the effective resonant frequency would be higher during the heating up temperatures than at a stable temperature. This results in a more positive τ_f because the temperature of the DR on top would be lower. Therefore, we thought the effect of different position of the upper DR on the lower DR is not obvious in our study as we measured τ_f after the temperature was stable.

CONCLUSIONS

ZN/ST stacked resonators with a zero τ_f have been obtained by adjusting the volume percentage of ST. f_{25} decreased from 9.27 GHz for 8.57% ST to 4.94 GHz for 27.27% ST. f_{25} decreased ~0.232 GHz on average for 1% ST increase. ε_r increased from 24.6 for 8.57% ST to 66.2 for 27.27% ST. ε_r increased ~2.22 on average for 1% ST increase. $Q \times f$ decreased in a different tendency for lower

(< 15.79%) and higher (> 15.79%) volume percentage of ST. $Q \times f$ rapidly decreased from 48,230 GHz for 8.57% ST to 7798 GHz for 15.79% ST and then 962 GHz for 27.27% ST. $Q \times f$ decreased ~2530 GHz on average for 1% ST increase between 8.57% and 27.27% ST. A temperature-stable ZN/ST stacked resonator with τ_f = 0 ppm/°C is obtained for 8.57% ST. τ_f increased to 678 ppm/°C for 27.27% ST (~36.25 ppm/°C on average for 1% ST increase). τ_f increased in a different tendency for lower (< 15.79%) and higher (> 15.79%) volume percentage of ST. Dielectric properties: ε_r = 24.6, $Q \times f$ = 48230 GHz and τ_f = 0 ppm/°C are obtained for ZN/ST stacked resonator with 8.57% volume percentage of ST.

REFERENCES

1. W. Wersing, *Curr. Opin. Solid State Mater. Sci.* 1, 715 (1996).
2. M.T. Sebastian, I.N. Jawahar, and P. Mohanan, *Mater. Sci. Eng., B* 97, 258 (2003).
3. N.D. Kataria, I.N. Jawahar, and M.T. Sebastian, *Mater. Lett.* 60, 2642 (2006).
4. L. Li, X.M. Chen, and X.C. Fan, *J. Am. Ceram. Soc.* 89, 557 (2006).
5. C. Tsironis and V. Panker, *IEEE MTT* 31, 312 (1983).
6. J. Breeze, S.J. Penn, M. Poole, and N. McAlford, *Electron. Lett.* 36, 883 (2000).
7. X.M. Chen, L. Li, and X.Q. Liu, *Mater. Sci. Eng., B* 99, 255 (2003).
8. L. Li and X.M. Chen, *J. Am. Ceram. Soc.* 89, 2514 (2006).
9. L. Li, X.M. Chen, and X.C. Fan, *J. Eur. Ceram. Soc.* 26, 2817 (2006).
10. C.W. Kuo, *Thesis for Master of Science (in Chinese)* (Tainan: Department of Electronic Engineering, Kun Shan University, 2013).
11. D. Zhou, H. Wang, and X. Yao, *Mater. Chem. Phys.* 105, 151 (2007).
12. H.J. Lee, I.T. Kim, and K.S. Hong, *Jpn. J. Appl. Phys.* 36, L1318 (1997).
13. P.H. Sun, T. Nakamura, Y.J. Shan, Y. Inaguma, and M. Itoh, *Jpn. J. Appl. Phys.* 37, 5625 (1998).
14. Y.C. Liou and Y.L. Sung, *Ceram. Int.* 34, 371 (2008).
15. Y.L. Sung, C.Y. Liu, M.H. Weng, and Y.C. Liou, *Proceedings of 2005 MRS-T Annual Meeting* (in Chinese), Tamkang University, Taipei, Taiwan, November 25–26, 2005.
16. Y.C. Liou, C.T. Wu, and T.C. Chung, *J. Mater. Sci.* 42, 3580 (2007).
17. B.W. Hakki and P.D. Coleman, *I.E.E.E. Trans. Microw Theory Tech.* 8, 402 (1960).
18. W. Wersing, *Electronic Ceramics*, ed. B.C.H. Steele (London: Elsevier Applied Science, 1991), pp. 67–119.
19. W.W. Cho, K. Kakimoto, and H. Ohsato, *Jpn. J. Appl. Phys.* 43, 6221 (2004).