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Characterization of $MgTiO_3$–$CaTiO_3$-Layered Microwave Dielectric Resonators with $TE_{01\delta}$ Mode

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$MgTiO_3$ and $CaTiO_3$ ceramics were stacked in different schemes as the key components of the microwave dielectric resonators, and the microwave dielectric characteristics were evaluated with $TE_{01\delta}$ resonant mode. With increasing thickness fraction of $CaTiO_3$, the resonant frequency and $Q \times f$ value decreased, while the effective dielectric constant and temperature coefficient of resonant frequency increased. The microwave dielectric characteristics were also affected by the stacking scheme. These effects were analyzed by the finite-element method, and the predicted characteristics agreed well with the experimental results. It was indicated that the temperature-stable resonators with high-$Q \times f$ values could be attained by changing the thickness fractions of $CaTiO_3$, and the corresponding microwave dielectric characteristics could also be predicted by the finite-element method.

I. Introduction

Dielectric ceramics have been widely used for microwave applications as the key components of the dielectric resonators in the microwave circuits.[1] To attain miniaturized, high-Q and temperature-stable resonators, the microwave dielectric ceramics should have high dielectric constant (ε_r), high-$Q \times f$ value, and near-zero temperature coefficient of resonant frequency (τ_f).[1,2] Many ceramics have high-dielectric constants and high-$Q \times f$ values, although their temperature coefficients of resonant frequency are large and need to be tuned. Conventional tuning methods focus on mixing two materials with opposite temperature coefficients to form a solid solution.[3–6] However, not all ceramics can be tuned by this method as the possible incompatibility of ionic radius, ionic charge, or crystal structure will bring undesired secondary phases and damage the $Q \times f$ value much.

Modifying the resonator structure is another way to improve the temperature stability of the dielectric resonator.[7–9] Layered dielectric resonator is just such a new method by stacking two dielectric ceramics with opposite temperature coefficients of resonant frequency, and the near-zero temperature coefficient could be obtained without damaging the high-$Q \times f$ value much.[9–13] This method makes it possible to use ceramics with large temperature coefficients in temperature-stable resonators. Although much work on layered dielectric resonator has been done, many problems have not been understood deeply yet, such as the effects of the thickness ratio and stacking scheme. Accurate prediction of the microwave dielectric characteristics is also an important issue for the layered resonators.

In the present work, $MgTiO_3$ and $CaTiO_3$ ceramics are selected as the components of the layered resonators, as their microwave dielectric characteristics vary much from each other[14] so that the effects of the thickness ratio and stacking scheme are easily observed. (The measured properties in the present experiments are: $\varepsilon_r = 17.17$, $Q \times f = 92\,000$ GHz, $\tau_f = -50.0$ ppm/°C for $MgTiO_3$ and $\varepsilon_r = 174.3$, $Q \times f = 11\,260$ GHz, $\tau_f = 804.1$ ppm/°C for $CaTiO_3$). The effects of the thickness fraction of $CaTiO_3$ and the stacking scheme on the microwave dielectric characteristics for the layered resonators with $TE_{01\delta}$ mode are investigated and discussed in the present work. Moreover, the finite-element method is used for evaluating the layered dielectric resonators, and the predicted characteristics are compared with the experimental results.

II. Experimental Procedure

$MgTiO_3$ and $CaTiO_3$ powders were synthesized by a solid-state reaction process. MgO (97%), $CaCO_3$ (99%) and TiO_2 (99.5%) raw powders with proper ratios were mixed by ball milling in distilled water with zirconia media for 24 h and then calcined at 1100°C in air for 3 h to synthesize $MgTiO_3$ and $CaTiO_3$, respectively. $MgTiO_3$ and $CaTiO_3$ powders with organic binder of PVA water solution were pressed into cylindrical compacts and sintered at 1350°C in air for 3 h. XRD analysis was conducted to determine the phase constitutions of the as-prepared $MgTiO_3$ and $CaTiO_3$ ceramics. The as-sintered pellets with the same diameter of 10.60 mm were thinned to different thicknesses as desired, and polished well with paralleling and smooth surfaces. The pellets were stacked together carefully with different stacking schemes and thickness ratios, then they were placed in the shielded cavity with axial symmetry and could work as dielectric resonators. The total thickness for all these stacks was 5.00 ± 0.01 mm. As shown in Fig. 1, bi-layer and tri-layer resonators were investigated with four $MgTiO_3$–$CaTiO_3$ stacking schemes in the present experiment, and they were indicated by $MgTiO_3/CaTiO_3$, $CaTiO_3/MgTiO_3$, $MgTiO_3/CaTiO_3/MgTiO_3$, and $CaTiO_3/MgTiO_3/CaTiO_3$, respectively. The thickness of each layer and the thickness fractions of $CaTiO_3$ for these stacks are shown in Table I.

The resonant frequency and temperature coefficient of resonant frequency with $TE_{01\delta}$ mode between 20° and 80°C were determined by network analyzer (8720 ES, Agilent Technologies Inc., Palo Alto, CA) using the reflection-type cavity method, and the cross-section of the resonator is shown in Fig. 2(a). The unloaded $Q \times f$ value of the stack is calculated by fitting the measured complex reflection coefficients at different frequencies near the resonant frequency to a circle using a personal computer,[15] and then removing the effect of the loss of the cavity wall.[16] Assuming a homogeneous dielectric with the same diameter and thickness as the stack, if they give the same resonant frequency under the present conditions, the dielectric constant of the homogeneous dielectric is defined as the effective dielectric constant ($\varepsilon_{r,eff}$) of the stack. $\varepsilon_{r,eff}$ can be calculated with high accuracy using the finite-element method from the resonant frequency and dimensions of the stack.[17]

III. Finite-Element Analysis

The axis symmetry exists for $TE_{01\delta}$ mode and the electric field has only a rotational component E_θ that is also axis symmetri-

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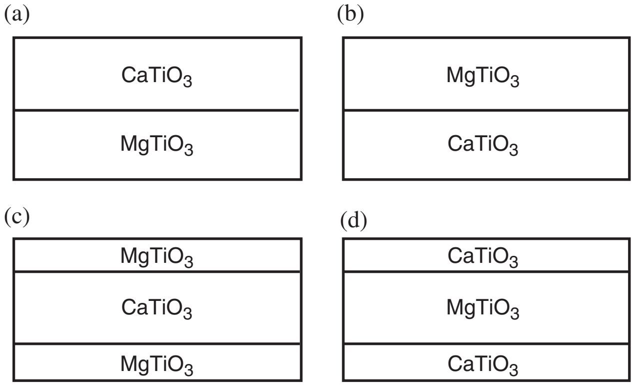

Fig. 1. Stacking schemes for the $MgTiO_3$–$CaTiO_3$ stacks, (a) $MgTiO_3$/$CaTiO_3$, (b) $CaTiO_3$/$MgTiO_3$, (c) $MgTiO_3$/$CaTiO_3$/$MgTiO_3$, and (d) $CaTiO_3$/ $MgTiO_3$/$CaTiO_3$.

cal, so the two-dimension finite-element method can be used to analyze the layered dielectric resonators, and only half of the cross-section needs to be analyzed. The half section for finite-element analysis is shown in Fig. 2(b), where the triangular first-order element is employed. Many resonant frequencies corresponding to $TE_{0n(p+\delta)}$ modes can be attained after resolving special linear equations, and the lowest is the resonant frequency of the layered resonator with $TE_{01\delta}$ mode. Also, the relative electric field intensity of each node can be given. The detailed analyzing process has been reported in the previous work.[18,19]

From the electric field distribution, the electric filling factor of each component, $P_{e,i}$, can be calculated by

$$P_{e,i} = \frac{1/2\int_V \varepsilon_{r,i}\vec{E}\cdot\vec{E}^{*}\mathrm{d}V}{\sum_i\left(1/2\int_V \varepsilon_{r,i}\vec{E}\cdot\vec{E}^{*}\mathrm{d}V\right)} = \frac{\int_S \varepsilon_{r,i}E_\theta^2\cdot r\mathrm{d}S}{\sum_i\left(\int_S \varepsilon_{r,i}E_\theta^2\cdot r\mathrm{d}S\right)} \quad (1)$$

For the layered resonators with $TE_{01\delta}$ mode, Alford *et al.*[10,11] have shown that the temperature coefficient of resonant frequency obeys

$$\tau_f = A + \sum_i (\tau_{f,i}P_{e,i}) \quad (2)$$

where $\tau_{f,i}$ is the temperature coefficient of resonant frequency for each component with the electric energy filling factor of unity and A is the effect of the cavity on τ_f because of the thermal expansion coefficient of the cavity wall (17 ppm/°C for Cu). A can be calculated by Kajfez's incremental frequency rule[16], and it is usually minus several tenths ppm/°C for the present resonators.

For the $MgTiO_3$–$CaTiO_3$ stack and individual $MgTiO_3$ and $CaTiO_3$ ceramics,

$$\tan\delta = \frac{f_0}{Q\times f},\ (\tan\delta)_i = \frac{f_0}{(Q\times f)_i} \quad (3)$$

and

$$\tan\delta = \frac{\sum_i((\tan\delta)_i P_{e,i})}{\sum_i P_{e,i}} \quad (4)$$

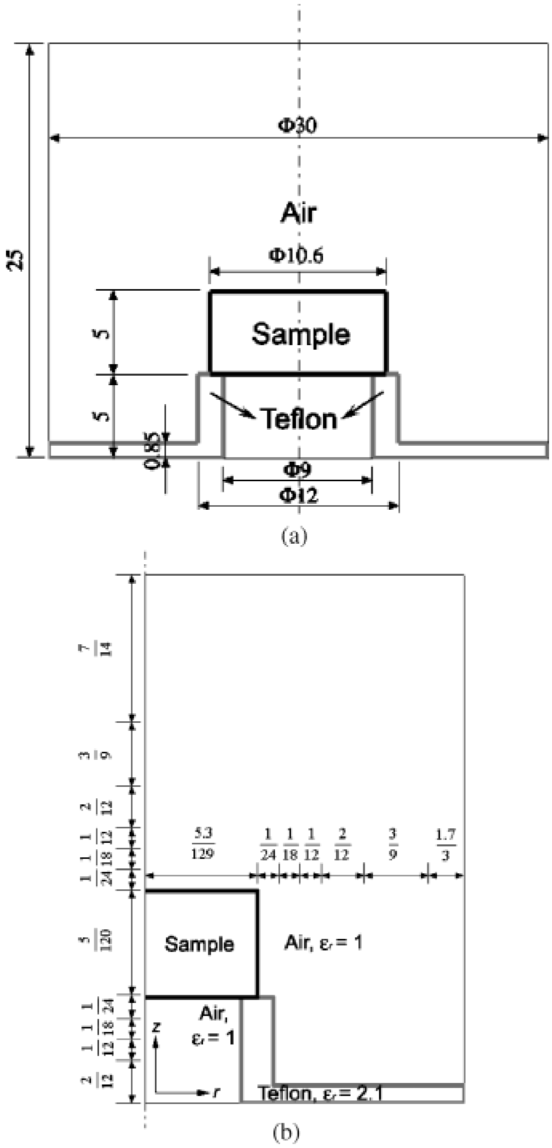

Fig. 2. (a) Structure of the resonator, (b) half of the cross-section of the resonator for finite-element analysis, where the fractional number n/m denotes that the length is n millimeter and divided into m segments averagely.

so we can attain

$$Q\times f = \left(\sum_i \frac{P_{e,i}}{(Q\times f)_i}\right)^{-1}\cdot\sum_i P_{e,i} \quad (5)$$

IV. Results and Discussion

Figure 3 shows XRD patterns of the as-prepared ceramics. Pure $CaTiO_3$ is attained in the present work, while a small amount of $MgTi_2O_5$ secondary phase is found in the $MgTiO_3$ ceramics. According to the previous reports on $MgTiO_3$-based ceramics, the $MgTi_2O_5$ secondary phase is difficult to eliminate completely

Table I. Experimental Parameters for the $MgTiO_3$–$CaTiO_3$ Stacks

Stacking scheme	Thickness of each layer (mm)				
$MgTiO_3$/$CaTiO_3$	3.75/1.25	3.33/1.67	2.50/2.50	1.67/3.33	1.25/3.75
$CaTiO_3$/$MgTiO_3$	1.25/3.75	1.67/3.33	2.50/2.50	3.33/1.67	3.75/1.25
$MgTiO_3$/$CaTiO_3$/$MgTiO_3$	1.88/1.25/1.88	1.67/1.67/1.67	1.25/2.50/1.25	0.83/3.33/0.83	0.62/3.75/0.62
$CaTiO_3$/$MgTiO_3$/$CaTiO_3$	0.62/3.75/0.62	0.83/3.33/0.83	1.25/2.50/1.25	1.67/1.67/1.67	1.88/1.25/1.88
Thickness fraction of $CaTiO_3$	0.25	0.33	0.50	0.67	0.75

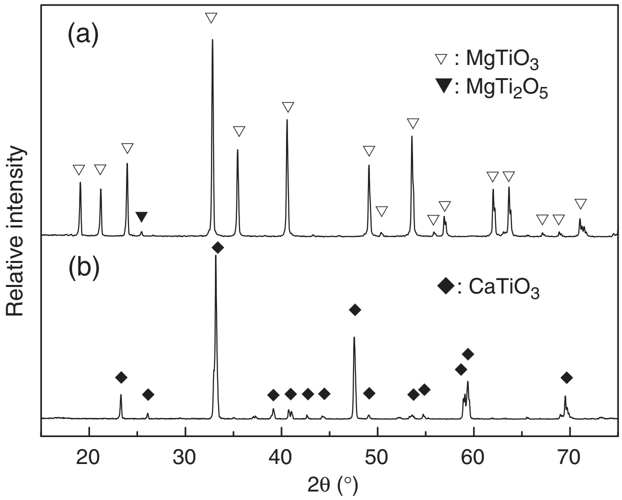

Fig. 3. XRD patterns of the as-prepared $MgTiO_3$ and $CaTiO_3$ ceramics: (a) $MgTiO_3$; (b) $CaTiO_3$.

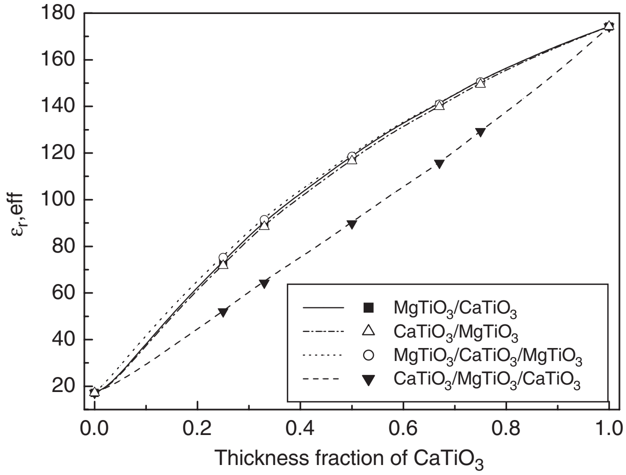

Fig. 5. Effective dielectric constant as function of thickness fraction of $CaTiO_3$ (lines, predicted; dots, experimental).

and it will damage the $Q \times f$ value.[20,21] But it does not matter much as the main aim of the present work is to discuss the mechanisms of the layered resonator. Surely, it is necessary to improve the preparing conditions to enhance the $Q \times f$ value for further research on resonators with high-$Q \times f$ value and temperature stability.

Figures 4–6 show the experimental and predicted resonant frequency, effective dielectric constant and temperature coefficient of resonant frequency for the $MgTiO_3$–$CaTiO_3$ layered resonators with $TE_{01\delta}$ mode as a function of the thickness fraction of $CaTiO_3$. The dots and lines correspond to the experimental and predicted values, respectively. To judge the accuracy of the finite-element method, the relative error is given as the difference between the predicted and experimental result divided by the experimental result. The relative errors for the resonant frequency, effective dielectric constant, and temperature coefficient of resonant frequency are within -0.54% to -0.01%, 0.02%–1.10%, and -0.76% to 0.68%, respectively. It is concluded that the finite-element method can give accurate predictions for the resonant frequency, effective dielectric constant and temperature coefficient of resonant frequency.

The thickness fraction of $CaTiO_3$ has a significant effect on the microwave properties for the layered resonators. This is because increasing the thickness fraction of $CaTiO_3$ means that $CaTiO_3$ contributes more and $MgTiO_3$ contributes less to the final microwave dielectric characteristics for the layered resonators with the same stacking scheme. Also, $CaTiO_3$ has a much higher dielectric constant and temperature coefficient of resonant frequency than $MgTiO_3$, so the resonant frequency decreases with increasing thickness fraction of $CaTiO_3$, while the effective dielectric constant and temperature coefficient of resonant frequency increase for the same stacking scheme.

The stacking scheme also affects the microwave dielectric characteristics as well as the thickness fraction of $CaTiO_3$. The curves of resonant frequency, effective dielectric constant, and temperature coefficient of resonant frequency as functions of the thickness fraction of $CaTiO_3$ for $CaTiO_3/MgTiO_3$ are very near to those for $MgTiO_3/CaTiO_3$, but they do not coincide. The differences are because of the unsymmetrical structure of the present cavity along the z direction (see Fig. 2(a)). The unsymmetrical structure leads to the difference of the electric field distribution between the $CaTiO_3/MgTiO_3$ and $MgTiO_3/CaTiO_3$ stacking schemes. Also, the dielectric constants of the air and supporter are much smaller than those of $MgTiO_3$ and $CaTiO_3$, so the effect of the unsymmetrical structure is slight. For example, the experimental resonant frequency of the $CaTiO_3/MgTiO_3$ resonator is 6–25 MHz larger than that for $MgTiO_3/CaTiO_3$ with the same thickness fraction of $CaTiO_3$. The difference is slight, but larger than the experimental error. To further understand the differences, the contours of the relative electric field intensity near the stacks for the $MgTiO_3/CaTiO_3$ and $CaTiO_3/MgTiO_3$ resonators with the same thickness fraction of $CaTiO_3$ of 0.5 are given in Figs. 7(a) and (b). The electric field distributions in $MgTiO_3$ and $CaTiO_3$ layers for bi-layer $CaTiO_3/MgTiO_3$ and $MgTiO_3/CaTiO_3$ resonators are actually a little different.

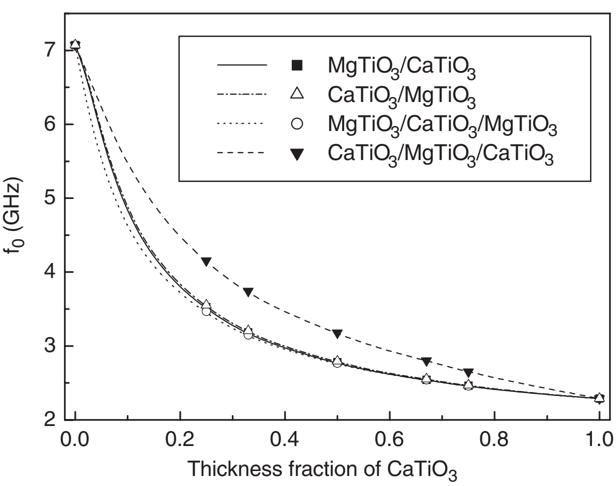

Fig. 4. Resonant frequency as a function of thickness fraction of $CaTiO_3$ (lines, predicted; dots, experimental).

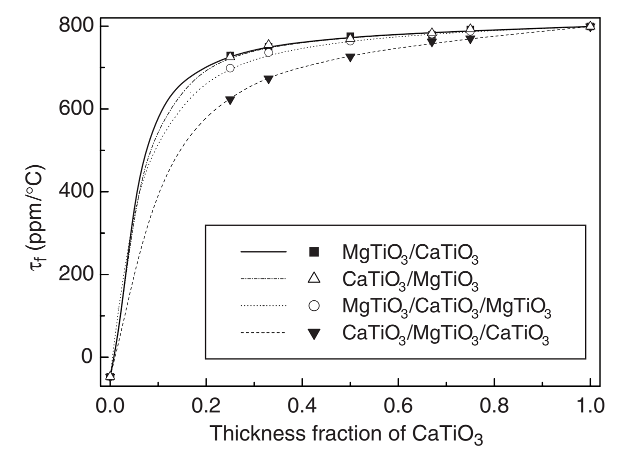

Fig. 6. Temperature coefficient of resonant frequency as a function of thickness fraction of $CaTiO_3$ (lines, predicted; dots, experimental).

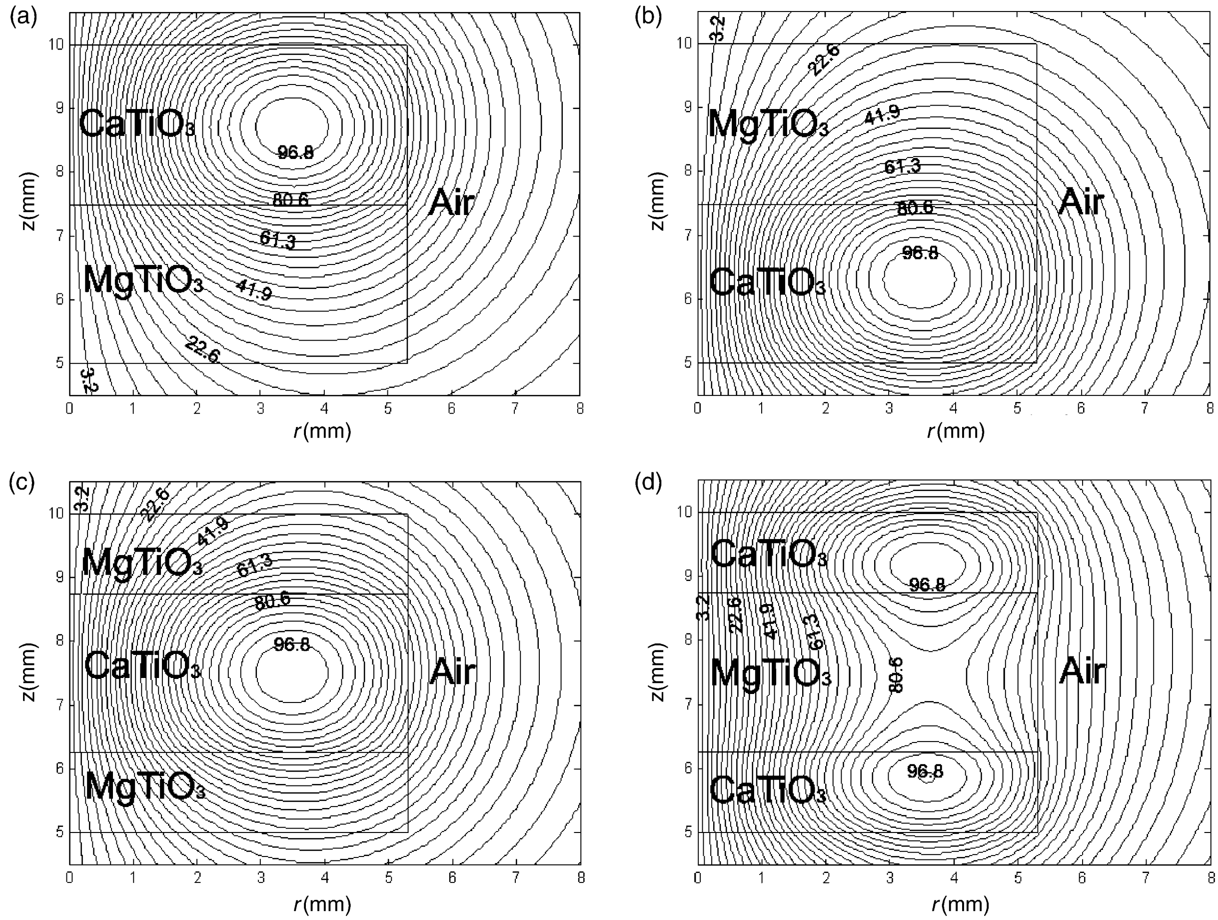

Fig. 7. Contours of the electric field intensity for the layered $MgTiO_3$–$CaTiO_3$ resonators with the same thickness fraction of $CaTiO_3$ of 0.5 with $TE_{01\delta}$ mode, simulated by the finite-element method: (a) $MgTiO_3/CaTiO_3$; (b) $CaTiO_3/MgTiO_3$; (c) $MgTiO_3/CaTiO_3/MgTiO_3$; and (d) $CaTiO_3/MgTiO_3/CaTiO_3$.

Among the tri-layer resonators, the curves for the resonators with a stacking scheme of $CaTiO_3/MgTiO_3/CaTiO_3$ deviate much more from the bi-layer resonators than those for $MgTiO_3/CaTiO_3/MgTiO_3$. A simple explanation is given from the structure similarity of the resonators as the microwave dielectric characteristics are determined by the components of the resonators and the schemes of the components. The air and the supporter (Teflon) have low dielectric constants of 1 and 2.1, respectively, and are designated as "L". Also, $MgTiO_3$ (with middle ε_r of 17.17) and $CaTiO_3$ (with high ε_r of 174.3) are designated as "M" and "H," respectively. Thus, according to the sequences of the dielectrics from the bottom to the top, the resonators with different schemes can be denoted as the following:

$MgTiO_3/CaTiO_3$: L–M–H–L,
$CaTiO_3/MgTiO_3$: L–H–M–L,
$MgTiO_3/CaTiO_3/MgTiO_3$: L–M–H–M–L, and
$CaTiO_3/MgTiO_3/CaTiO_3$: L–H–M–H–L.

It is obvious that the resonator with a stacking scheme of $MgTiO_3/CaTiO_3/MgTiO_3$ has a more similar structure to the bi-layer resonators than to that for $CaTiO_3/MgTiO_3/CaTiO_3$. Also, the contours of the relative electric field intensity give the same results (see Fig. 7).

The measured $Q \times f$ value of the $MgTiO_3$–$CaTiO_3$ stack as a function of the thickness fraction of $CaTiO_3$ is shown in Fig. 8(a), as the dots indicate. For the same stacking scheme, the $Q \times f$ value decreases with increasing the thickness fraction of $CaTiO_3$. The stacking scheme also has an effect on the $Q \times f$ value. For the same thickness fraction of $CaTiO_3$, the highest $Q \times f$ value is obtained for the stacking scheme of $CaTiO_3/MgTiO_3/CaTiO_3$. It is followed by $MgTiO_3/CaTiO_3/MgTiO_3$, and the bi-layer $MgTiO_3/CaTiO_3$ and $CaTiO_3/MgTiO_3$ resonators indicate the lowest $Q \times f$ values which are near the same for both cases. The predicted $Q \times f$ values are given in Fig. 8(a), as the lines indicate. The relative error between the predicted and measured $Q \times f$ value $(((Q \times f)_{pre} - (Q \times f)_{meas})/(Q \times f)_{meas})$ is from -1.4% to 4.7%, depending on the stacking scheme and thickness fraction of $CaTiO_3$. Although the prediction is not very accurate, the predicted trends are consistent with the experimental results. The errors may be attributed to many uncertainties of the $Q \times f$ measurement, such as difference between the samples,[12] limited accuracy of the network analyzer,[22] imperfect data fitting to an ideal circle,[22] effect of the coupling coefficient[23] and so on. For each stacking scheme, a sharp rise of $Q \times f$ value is observed when the thickness fraction of $CaTiO_3$ decreases to a certain value (see Fig. 8(b)).

The layered resonators with zero temperature coefficient of resonant frequency can be attained since $MgTiO_3$ and $CaTiO_3$ have reverse temperature coefficients of resonant frequency. The finite-element method gives the predicted thickness fractions of $CaTiO_3$ and corresponding microwave dielectric characteristics for the temperature-stable-layered dielectric resonators, as shown in Table II, and high-$Q \times f$ values can be attained. For different stacking schemes, the zero temperature coefficient of resonant frequency is attended at different thickness fractions of $CaTiO_3$, where the dielectric constant and $Q \times f$ value are almost the same for all these situations. This is because the effects of $CaTiO_3$ on the temperature coefficient of resonant frequency are dependent on the stacking schemes. Although the predicted microwave dielectric properties of the temperature-stable-layered resonators are not better than those of the (Mg,Ca)TiO_3-based resonators ($\varepsilon_r = 20$–22, $Q \times f = 56\,000$–$92\,000$ GHz and $\tau_f \approx 0$ ppm/°C[21–26]), they are expected to be much improved through optimizing the preparing conditions to improve the corresponding properties, especially the $Q \times f$ values of $MgTiO_3$ and $CaTiO_3$.

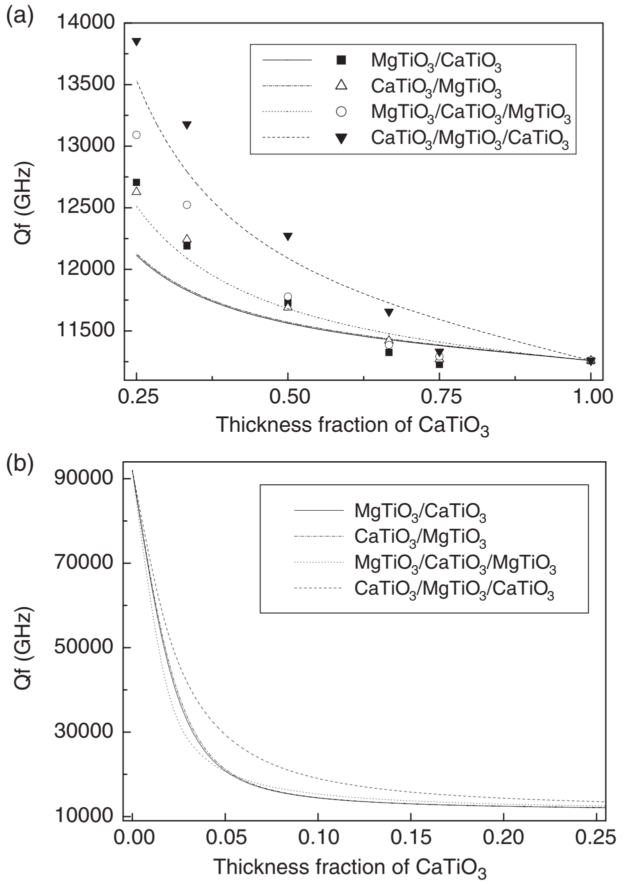

Fig. 8. $Q \times f$ value as a function of thickness fraction of $CaTiO_3$ (lines, predicted; dots, experimental).

Table II. Predicted Temperature-Stable Layered $MgTiO_3$–$CaTiO_3$ Resonators with Different Stacking Schemes

Stacking scheme	Thickness fraction of $CaTiO_3$	f_0 (GHz)	$\varepsilon_{r,eff}$	$Q \times f$ (GHz)	τ_f (ppm/°C)
$MgTiO_3/CaTiO_3$	0.0085	6.9135	18.03	66,850	0
$CaTiO_3/MgTiO_3$	0.0090	6.9149	18.02	66,870	0
$MgTiO_3/CaTiO_3/MgTiO_3$	0.0047	6.8978	18.12	66,880	0
$CaTiO_3/MgTiO_3/CaTiO_3$	0.0103	6.9019	18.10	66,860	0

Although the as-prepared layered dielectric resonators may have high $Q \times f$ and near-zero temperature coefficient of resonant frequency, they are hard to handle for practical applications as the different layers are separate. To resolve this problem, adhesive with low dielectric loss should be used to bond the different layers. Surely, the adhesive will have an effect on the microwave dielectric characteristics of the layered resonators, and this effect will be investigated in further work.

V. Conclusion

The microwave dielectric characteristics for the layered $MgTiO_3$–$CaTiO_3$ resonators with $TE_{01\delta}$ mode are discussed in detail. The stacking scheme has a significant effect on the electric field distribution as well as the thickness fraction of $CaTiO_3$. The finite-element method can give accurate predictions for the microwave dielectric characteristics, and it is a useful tool for analyzing the layered dielectric resonators and designing temperature-stable-layered dielectric resonators. Temperature-stable dielectric resonators with high-$Q \times f$ values are expected to be attained through this new method, and the details should be investigated further in the future work.

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