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Frequency-dependent Qf value of low-loss $Ba_2Ti_9O_{20}$ ceramics at microwave frequencies

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Abstract

The microwave dielectric properties of low-loss $Ba_2Ti_9O_{20}$ ceramics were evaluated by the resonant cavity method, and the frequency dependence of Qf value was investigated. TE_{011} resonant mode was used for measurement, and the measurement frequency was changed by adjusting the sample size. The measurement frequency could also be tuned over a wide range by using higher-order TE_{0np} modes for a sample with a fixed size. The measured Qf value of $Ba_2Ti_9O_{20}$ ceramics increased significantly with increasing the frequency from 4.55 to 10.74 GHz. The result conflicted with the common recognition that the Qf value of microwave dielectric ceramics was a frequency-independent constant at microwave frequencies. The frequency dependence of Qf value was attributed to the extrinsic dielectric loss, which originated from extrinsic factors such as grain boundary, oxygen vacancy and pore.

Keywords: Frequency dependence; Qf value; Microwave frequencies; Extrinsic dielectric loss

1. Introduction

Microwave dielectric ceramics are characterized by the low dielectric loss at microwave frequencies, and they have been widely used in microwave circuits as resonators, filters, etc. [1–3]. For a perfect ionic crystal, only the intrinsic factors including electronic and ionic polarizations contribute to the microwave dielectric properties. The dielectric constant originated from the electronic polarization is the optical frequency dielectric constant (ε_∞) at microwave frequencies, and the corresponding dielectric loss can be neglected. Furthermore, the ionic polarization can be described by several damped harmonic oscillators, which correspond to the lattice vibrations and usually resonate at far-infrared frequencies higher than 10^{12} Hz [1,4,5]. At microwave frequencies which are much lower than the resonant frequencies of the harmonic oscillators, the intrinsic complex dielectric constant and dielectric loss can be represented by

$$\varepsilon'_I = \varepsilon_\infty + \sum_j \frac{S_j}{\omega_j^2} \tag{1}$$

$$\varepsilon''_I = \sum_j \frac{\omega \gamma_j S_j}{\omega_j^4}, \tag{2}$$

and

$$\tan\delta_I = \frac{\varepsilon''_I}{\varepsilon'_I} = \frac{\omega \sum_j \gamma_j s_j / \omega_j^4}{\varepsilon_\infty + \sum_j s_j/\omega_j^2}, \tag{3}$$

where ω_j, S_j, and γ_j are the resonant angular frequencies, strength and damping constant of the damped harmonic oscillator, and ω is the angular frequency [1,4–7]. It is deduced that the real part of the dielectric constant is a constant at microwave frequencies, and the dielectric loss is proportional to the frequency. So the parameter Qf is usually used to describe the loss property of the microwave dielectric ceramics, which is defined by the product of Q value (reciprocal dielectric loss) and frequency. It has been widely accepted for decades that the Qf value is a frequency-independent constant at microwave frequencies for low-loss dielectric ceramics [1–3].

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It should be noted that the Qf value as a frequency-independent constant is based on the assumption that only ionic polarization contributes to the dielectric loss, which is valid for a perfect crystal. However, for practical microwave dielectric ceramics, the extrinsic factors such as grain boundary, oxygen vacancy and pore may also contribute to the dielectric loss. In fact, the Qf value of the microwave dielectric ceramics at a certain frequency is usually sensitive to the preparation conditions and microstructures [8,9], which indicates the contribution of extrinsic factors. The extrinsic factors may also affect the frequency dependence of Qf value, so the constant Qf value is doubtful for practical microwave dielectric ceramics. The Qf value as a function of frequency at microwave frequencies has attracted some attention, while distinguishing results have been reported by different researchers [1,3,10–15]. Moreover, the origin of the possible frequency dependence of Qf value is unclear. So in the present work, the frequency dependence of Qf value for a typical microwave dielectric ceramic ($Ba_2Ti_9O_{20}$) is investigated, and the origin of the frequency dependence is also discussed.

2. Experimental procedure

The $Ba_2Ti_9O_{20}$ ceramics with a diameter of about 10.88 mm and thickness of 1–5.5 mm were prepared by a standard solid state reaction method [16,17]. The dielectric constant and Qf value were measured by the resonant cavity method at room temperature. One-port cylindrical copper resonant cavity with smooth surfaces was pasted with silver and gold to decrease the loss of metal walls. TE_{0np} resonant modes were used to measure the dielectric properties [12], among which TE_{011} mode (also indicated by $TE_{01\delta}$ mode for the resonant cavity method) was the most widely used resonant mode [18]. The measurement frequency (also the resonant frequency) was changed by adjusting the sample thickness from 1 to 5.5 mm for TE_{011} mode. It was also tuned over a wide frequency range by using high-order TE_{0np} modes for the sample with the thickness 5.5 mm. The dielectric constant was calculated through an iterative process from the resonant frequency and sample size by finite element analysis [19,20], and the dielectric loss (tan δ) was obtained by [18]

$$\frac{1}{Q_L} = (1+\kappa)\left(P_e \tan\delta + \frac{1}{Q_c}\right), \quad (4)$$

where Q_L (loaded quality factor) and κ (coupling coefficient) could be calculated from the resonant curve by linear fractional curve fitting [21], P_e (electric energy filling factor of the sample) was calculated by modeling the electric field distribution through finite element analysis, [20] and Q_c (quality factor of the conductive metal wall) was obtained by the incremental frequency rule [22]. The Qf value is then calculated from the product of the resonant frequency and reciprocal dielectric loss.

3. Results and discussion

The measurement frequency can be tuned from 4.55 to 7.60 GHz by decreasing the sample thickness from 5.5 to 1 mm for TE_{011} mode (indicated by method 1), and it is up to 10.74 GHz by using high-order TE_{0np} modes (TE_{012}, TE_{021} and TE_{022}) for the sample with the thickness 5.5 mm (indicated by method 2). According to Eq. (4), the measurement accuracy for Qf value is dominated by P_e for a high-Q cavity. In the present work, the electric energy filling factor of the sample decreases from 0.952 to 0.765 with decreasing sample thickness from 5.5 to 1 mm for method 1, and it is between 0.682 and 0.972 for method 2. The electric energy filling factor is high enough to ensure the high measurement accuracy for Qf value. The dielectric properties have not been measured at higher frequencies for method 1, because further decrease in sample thickness causes a rapid decrease in P_e and unreliable measured Qf value. Furthermore, only TE_{011}, TE_{012}, TE_{021} and TE_{022} resonant modes are used for method 2 due to the increasing spurious resonant modes at higher frequencies.

As shown in Fig. 1, the measured dielectric constant of $Ba_2Ti_9O_{20}$ ceramics varies between 39.03 and 39.30 over the measurement frequency range for the two methods. Considering the measurement error, the dielectric constant is regarded to be independent of frequency. Whereas, the measured Qf value exhibits significant dependence on frequency, as shown in Fig. 2. It increases from 32,600 to 37,100 GHz with increasing frequency from 4.55 to 7.60 GHz for method 1. The increase in Qf value with frequency has been observed for some microwave dielectric ceramics when the measurement frequency is tuned by changing the sample size [1,3,11], and it is usually explained by the statistically increasing flaws and defects for the larger sample which resonates at lower frequency [3]. This explanation indicates that the Qf value is determined by the concentrations of the flaws and defects, and it should be a

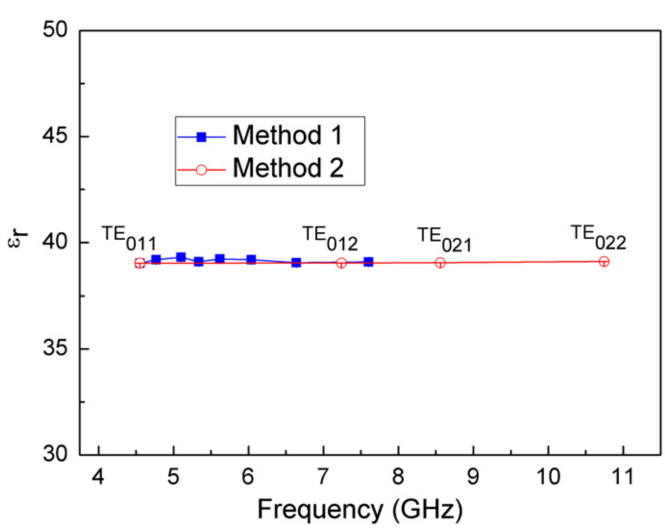

Fig. 1. Dielectric constant of $Ba_2Ti_9O_{20}$ ceramics as a function of frequency.

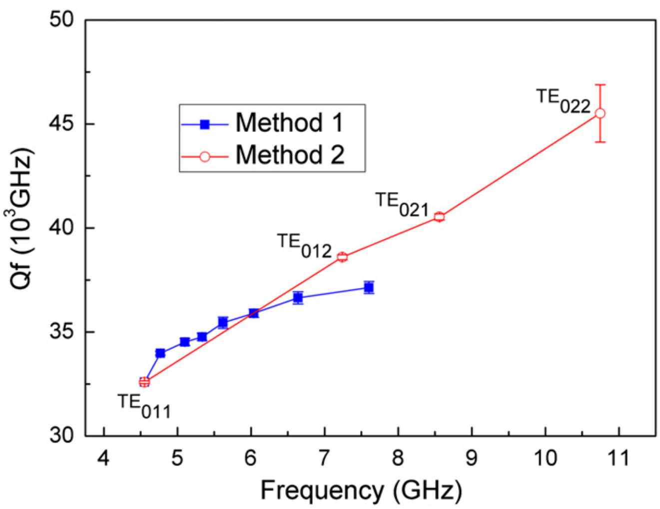

Fig. 2. Qf value of $Ba_2Ti_9O_{20}$ ceramics as a function of frequency.

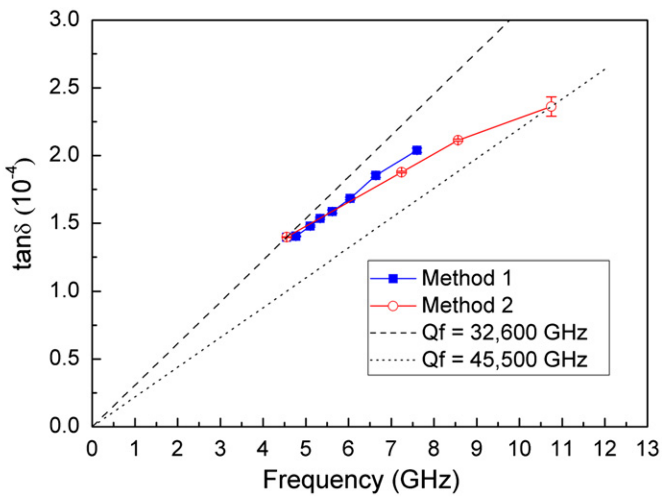

Fig. 3. Dielectric loss of $Ba_2Ti_9O_{20}$ ceramics as a function of frequency.

frequency-independent constant for a sample with a fixed size. However, the increase in Qf value is also observed for a sample with a fixed size. As shown in Fig. 2, when the measurement frequency increases up to 10.74 GHz for method 2, the Qf value of $Ba_2Ti_9O_{20}$ ceramics further increases to 45,500 GHz, which is about 1.4 times of that at 4.55 GHz. It is indicated that the above explanation is doubtful. Furthermore, the results of the two methods conflict with the common recognition for low-loss microwave dielectric ceramics that the Qf value is a frequency-independent constant at microwave frequencies [1–3].

The Qf value as a frequency-independent constant at microwave frequencies is based on the assumption that only the ionic polarization contributes to the dielectric loss for a perfect crystal [1,4,5]. The effect of ionic polarization on dielectric loss (intrinsic dielectric loss) is proportional to frequency, so that the Qf value is a frequency-independent constant (see Eq. (3)). However, the extrinsic factors including grain boundary, oxygen vacancy and pore always exist for practical ceramics, and they may also contribute to the microwave dielectric properties. The effect of extrinsic factors on the dielectric constant of low-loss microwave dielectric ceramics is usually slight and can be neglected, so the dielectric constant shows little frequency dependence over the measurement frequency range, as shown in Fig. 1. However, the dielectric loss of the microwave dielectric ceramics is very low and usually in the order of magnitude of 10^{-3}–10^{-5} [1–3], the effect of extrinsic factors on dielectric loss (extrinsic dielectric loss) may be significant. The overall dielectric loss of the microwave dielectric ceramics is composed of intrinsic dielectric loss ($\tan\delta_I$) and extrinsic dielectric loss ($\tan\delta_E$), so

$$\tan\delta = \tan\delta_E + \tan\delta_I = \tan\delta_E + f/\mathrm{Qf_I}, \quad (5)$$

where $\mathrm{Qf_I}$ represents the constant intrinsic Qf value. According to Eqs. (3) and (5), the Qf value should not be a frequency-independent constant if the extrinsic dielectric loss is not proportional to frequency. Fig. 3 shows the measured dielectric losses of the $Ba_2Ti_9O_{20}$ ceramics by methods 1 and 2 as functions of frequency, and the dash and point lines correspond to the constant Qf values with 32,600 and 45,500 GHz, respectively. The measured dielectric losses as functions of frequency derive significantly from the two lines that pass the origin point of the coordinate, which reflects the effect of the extrinsic dielectric loss.

The extrinsic dielectric loss as a function of frequency can be calculated by Eq. (5) from the measured dielectric loss and intrinsic Qf value, and the latter is usually obtained from the infrared reflection spectra for low-loss microwave dielectric ceramics [5–7]. However, the intrinsic Qf value of $Ba_2Ti_9O_{20}$ ceramics has not been reported due to the complex crystal structure [23]. So $\mathrm{Qf_I}$ is assumed to be a constant varying from 50,000 to 100,000 GHz in the present work, and the calculated extrinsic dielectric loss as a function of frequency for method 2 is shown in Fig. 4. The calculated extrinsic dielectric loss exhibits significant dependence on frequency. With increasing the frequency from 4.55 to 10.74 GHz, the calculated extrinsic dielectric loss decreases for $\mathrm{Qf_I} = 50{,}000$ GHz, while it increases for $\mathrm{Qf_I} = 100{,}000$ GHz. When $\mathrm{Qf_I}$ is between 60,000 and 80,000 GHz, the calculated extrinsic dielectric loss increases first and then decreases with frequency. The calculated extrinsic dielectric loss is not proportional to the frequency for all cases, which indicates that the extrinsic dielectric loss is responsible for the frequency dependence of Qf value. Moreover, the effects of extrinsic factors on the dielectric properties can be described by the dielectric relaxation, for which the dielectric loss increases first and reaches a peak value and then decreases with increasing the frequency [24]. It is indicated that the frequency dependence of the calculated extrinsic dielectric loss corresponds to the dielectric relaxation for each assumed $\mathrm{Qf_I}$ value, and the frequency where the calculated extrinsic dielectric loss reaches the peak value is dependent on $\mathrm{Qf_I}$.

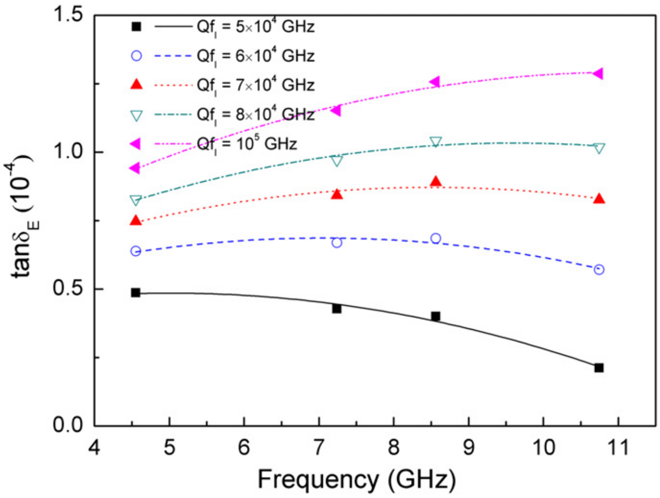

Fig. 4. Calculated extrinsic dielectric loss of $Ba_2Ti_9O_{20}$ ceramics for method 2 as a function of frequency with different assumed intrinsic Qf values.

Based on the above discussion, it is deduced that the Qf value for other microwave dielectric ceramics may also exhibits significant frequency dependence, since the extrinsic factors cannot be avoided for practical ceramics. Interestingly, different results for the frequency dependence of Qf value have been reported for $Ba(Mg_{1/3}Ta_{2/3})O_3$ microwave dielectric ceramics. Krupka et al. have observed that the Qf value of $Ba(Mg_{1/3}Ta_{2/3})O_3$ ceramics is nearly independent of frequency [12], while significant frequency dependence of Qf value is suggested in Ref. [15]. The distinguishing results may be due to the microstructural differences between the samples that lead to the different effects of extrinsic factors on Qf value and its frequency dependence. For practical microwave dielectric ceramics, attention should be paid to when using the Qf value to describe the loss property, since the arbitrary usage of the frequency-independent Qf value may bring an unreliable parameter at frequencies deviating from the measurement frequency. Furthermore, it is strongly suggested that the measurement frequency should be always noted when mentioning the Qf value.

4. Conclusions

In conclusion, the frequency dependence of Qf value for low-loss $Ba_2Ti_9O_{20}$ ceramics at microwave frequencies has been investigated by the resonant cavity method. The measured Qf value increases significantly with increasing frequency from 4.55 to 10.74 GHz, and it conflicts with the common recognition for low-loss microwave dielectric ceramics that the Qf value is a frequency-independent constant at microwave frequencies. The extrinsic dielectric loss is not proportional to the frequency, and it is responsible for the frequency dependence of Qf value. Attention should be paid when using the constant Qf value to describe the loss property, and it is strongly suggested that the measurement frequency should be always noted when mentioning the Qf value.

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