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Journal of the European Ceramic Society 26 (2006) 2165–2168

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Microwave dielectric properties of $BaTi_4O_9$ thin film

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Abstract

$BaTi_4O_9$ thin films have been prepared by RF magnetron sputtering on the $Pt/Ti/SiO_2/Si$ substrates and the dielectric properties of the $BaTi_4O_9$ film have been investigated at microwave frequency range. The homogeneous $BaTi_4O_9$ thin film was obtained when the film was grown at 550 °C and rapid thermal annealed (RTA) at 900 °C for 3 min. The circular-patch capacitor (CPC) was used to measure the microwave dielectric properties of the film. The dielectric constant (ε_r) and the dielectric loss ($\tan\delta$) were successfully measured up to 6 GHz. The ε_r of the $BaTi_4O_9$ thin film slightly increased with the increase of the film thickness. However, the $\tan\delta$ decreased with increasing the thickness of the film. The ε_r of $BaTi_4O_9$ thin film was similar to that of the $BaTi_4O_9$ ceramics, which is about 36–39. The $\tan\delta$ of the film with 460 nm thickness was very low approximately, 0.0001 at 1–3 GHz. Since $BaTi_4O_9$ film has a high ε_r and a low $\tan\delta$, the $BaTi_4O_9$ film can be used as the microwave devices.

Keywords: Films; Microstructure; Dielectric properties; Capacitors; $BaTi_4O_9$

1. Introduction

Recently, dielectric thin films have been intensively studied for the applications to the dynamic random access memory (DRAM) and the wireless-communication systems.[1,2] The required properties of such dielectric thin films are the high dielectric constant (ε_r) and the low dielectric loss ($\tan\delta$).[3] The operating frequencies of the both DRAM and wireless-communication systems range from hundreds of megahertz to tens of gigahertz. Therefore, the characterization of the dielectric thin films in the GHz range is essential to develop the devices operating at microwave frequencies. However, the dielectric properties of the dielectric thin film have mainly been studied in the kHz–MHz frequency range.[4]

The $BaO–TiO_2$ system has been extensively studied because it has many compounds, which have excellent microwave dielectric properties.[5,6] The $BaTi_4O_9$ ceramic shows good microwave dielectric properties of $\varepsilon_r = 36–39$, $Q = 2500–5000$ at 4–10 GHz and the temperature coefficient of the resonance frequency (τ_f) < 20 ppm/°C.[7,8] All the previous works on the $BaTi_4O_9$ system have been concentrated on the bulk ceramics but no research has been carried out on the structural and dielectric properties of the thin films. In this paper, the $BaTi_4O_9$ thin film was grown on the $Pt/Ti/SiO_2/Si$ substrate and the ε_r and $\tan\delta$ of the $BaTi_4O_9$ thin film were investigated in the microwave frequency range for the application to the microwave thin film devices.

2. Experimental procedure

A homogeneous $BaTi_4O_9$ thin film was successfully grown by RF sputtering method when the film was grown at 550 °C and rapid thermal annealed (RTA) at 900 °C for 3 min. The detailed growth conditions were reported in the other paper.[9] The microstructure of the $BaTi_4O_9$ thin film was analyzed using transmission electron microscopy (TEM:Hitachi H-9000NAR Ibaraki, Japan). The interface between the film and electrode was investigated by the auger electron spectroscopy (AES:Physical Electronics PHI 680 Auger Spectroscopy). To measure the dielectric properties of the $BaTi_4O_9$ thin film at microwave frequencies, a circular-patch capacitor (CPC) structure was used.[10] Al was deposited on the $BaTi_4O_9$ film and patterned by the one-step photolithography to form the top electrode of the circular-patch capacitor. The detailed shape and size of CPC is shown in Fig. 1(a). The reflection coefficient of incident microwaves which were used to calculate the impedance of the $BaTi_4O_9$ thin

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0955-2219/$ – see front matter
doi:10.1016/j.jeurceramsoc.2005.09.089

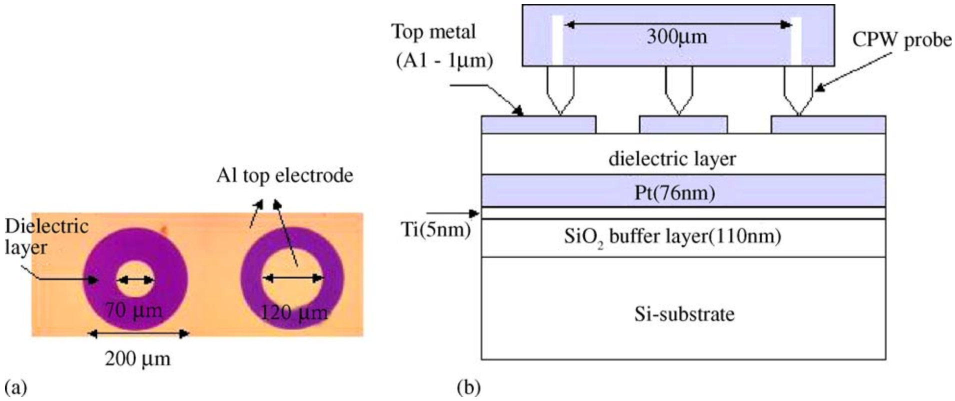

Fig. 1. Schematic diagrams of: (a) top view of the circular-patch capacitor; and (b) the cross-sectional view of the specimen with CPW probe.

film was measured by the vector network analyzer (VNA: HP 8710C) with a Be–Cu coplanar waveguide (CPW) probe (Picoprobe, USA). The schematic diagram of the cross-sectional view with the CPW probe is shown in Fig. 1(b).

3. Results and discussion

Fig. 2 shows a cross-sectional TEM bright field image of the $BaTi_4O_9$ thin film grown on $Pt/Ti/SiO_2/Si$ substrate at 550 °C and RTA at 900 °C for 3 min under O_2 atmosphere. The inset shows the electron diffraction pattern taken from the same area. The electron diffraction pattern was identified as the [0 1 0] zone axis electron diffraction pattern of the $BaTi_4O_9$ phase which has the orthorhombic structure with the lattice parameters of $a = 1.453$ nm, $b = 0.379$ nm and $c = 0.629$ nm.[11] The $BaTi_4O_9$ film with the thickness of 150 nm was uniformly formed and the interface between film and Pt substrate was relatively sharp. The interface between $BaTi_4O_9$ thin film and Pt electrode was also investigated using AES as shown in Fig. 3. Although the films were annealed at relatively high temperature, the diffusion of Pt ion into the thin film or that of Ba or Ti ions into the Pt electrode was insignificant. Therefore, it is considered that the interface between $BaTi_4O_9$ and Pt electrode is chemically sharp. Above results indicate that the homogeneous $BaTi_4O_9$ thin film was well developed on $Pt/Ti/SiO_2/Si$ substrate.

A CPC was used to measure the dielectric properties of the $BaTi_4O_9$ film at microwave frequency. The CPC is consisted of a ring-shape capacitor and an outer capacitor as shown in Fig. 1(a). It is impossible to calculate the impedance of the outer capacitor. Therefore, to obtain the measured impedance of the $BaTi_4O_9$ film (Z_{film}) without the effect of outer capacitor, the impedance of the CPC with a large inner diameter (Z_{a2}) was subtracted from that of CPC with a small inner diameter (Z_{a1}) as shown in following equation:

$$Z_{\mathrm{film}} = Z_{a1} - Z_{a2} = \frac{R_{\mathrm{sb}}}{2\pi} \ln\left(\frac{a_2}{a_1}\right) + \frac{1}{j\omega\pi}\frac{t}{\varepsilon_0\varepsilon_{\mathrm{r}}}\left(\frac{1}{a_1^2} - \frac{1}{a_2^2}\right) \quad (1)$$

where a_1 and a_2 are the diameters of inner capacitor of the each CPC, R_{sb} is the sheet resistance of the Pt bottom electrode and ε_0 the permittivity of free space (8.854×10^{-12} F/m), t the thickness of the thin film, ω the angular frequency and ε_{r} the complex dielectric constant of $BaTi_4O_9$ film.[10] The ε_{r} is then easily calculated from Eq. (1) when Z_{a1} and Z_{a2} were measured. The measured $\tan\delta$ ($\tan\delta_{\mathrm{m}}$) of the $BaTi_4O_9$ film is the ratio of the real and imaginary parts of the Z_{film}. Fig. 4 shows the $\tan\delta_{\mathrm{m}}$ of the $BaTi_4O_9$ film and it linearly increased with the

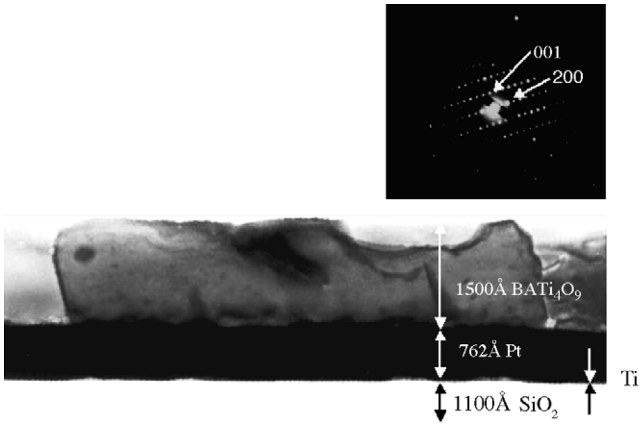

Fig. 2. Cross-sectional TEM bright field image of the $BaTi_4O_9$ thin film grown at 550 °C for 1 h and rapidly thermal annealed at 900 °C for 3 min.

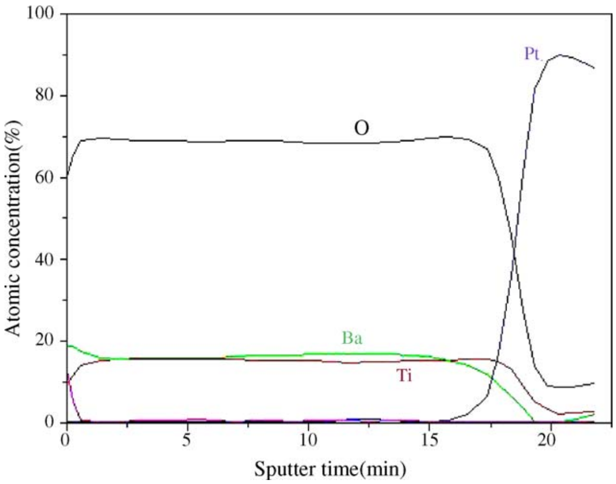

Fig. 3. Auger electron spectroscopy of the $BaTi_4O_9$ thin film grown at 550 °C for 1 h and rapidly thermal annealed at 900 °C for 3 min.

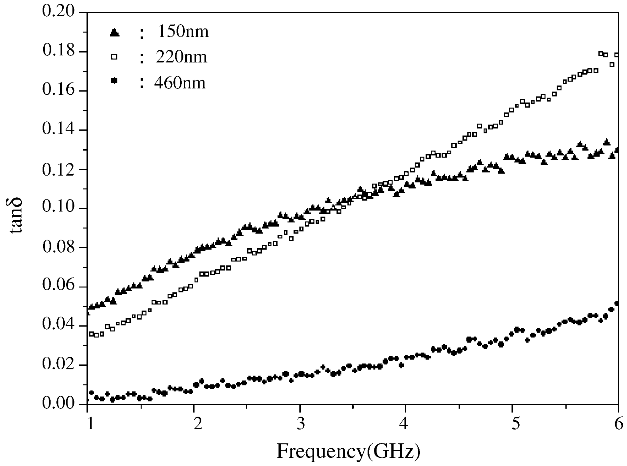

Fig. 4. The measured tan δ of the $BaTi_4O_9$ thin films with various thicknesses as a function of frequency.

increase of the frequency. The Z_{film} has the additional series resistances, which were caused by resistances in the top/bottom electrodes and the contact resistance between the CPW probe and inner/outer top electrodes. Therefore, the tan δ_m shown in Fig. 4 also has the additional losses, which caused the increase of the tan δ_m with the increase of the frequency. The resistances of the top/bottom electrodes are constant, thus their effects on the Z_{film} can be negligible. On the other hand, the contact resistance changes in each measurement. Therefore, the equivalent-circuit model shown in Fig. 5 is generally used to obtain the tan δ of pure dielectrics (tan δ_d) without an additional loss caused by the contact resistance.[12]The series resistance in this equivalent-circuit model represents the contact resistance. Therefore, the impedance of the equivalent-circuit, which is the same as Z_{film} can be expressed in the following equation,

$$Z_{\mathrm{film}} = \frac{R_p}{1 + j\omega C_p R_p} + R_s = R_p(\tan\delta_d)^2 + R_s - j\frac{1}{\omega C_p} \qquad (2)$$

where C_p is the capacitance, R_p the intrinsic resistance of the dielectric thin film, and R_s the series resistance due to contact resistance between the CPW probe and top electrode. Moreover, the tan δ_d and $Re(Z_{\mathrm{film}})$ of the dielectrics can be expressed as the following equations:

$$\tan\delta_d = \frac{Re(Z_{\mathrm{film}}) - R_s}{Im(Z_{\mathrm{film}})} \qquad (3)$$

$$Re(Z_{\mathrm{film}}) = Im(Z_{\mathrm{film}}) \times \tan\delta_d + R_s = -\frac{1}{\omega C_p} \times \tan\delta_d + R_s \qquad (4)$$

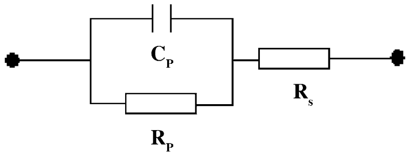

Fig. 5. Equivalent-circuit model for the circular-patch capacitor.

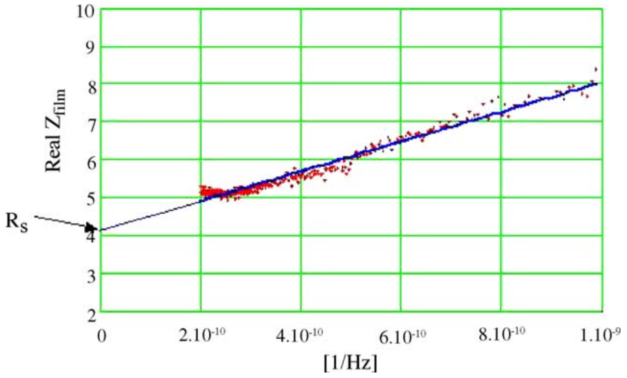

Fig. 6. Real part of the measured impedance of the $BaTi_4O_9$ thin film (Z_{film}) containing contact resistance.

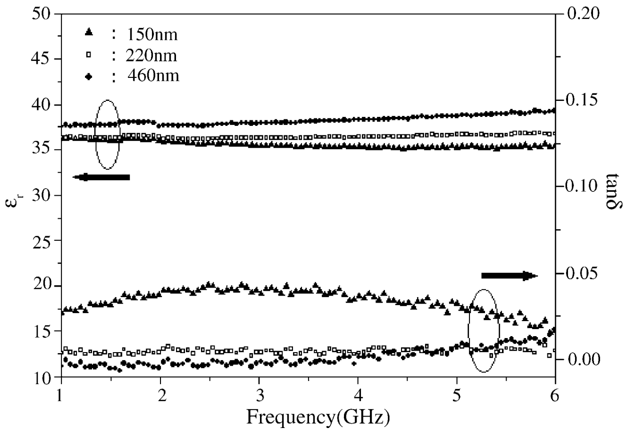

Fig. 7. The ε_r and tan δ of the $BaTi_4O_9$ thin films with various thicknesses as a function of frequency.

As shown in Eq. (4), R_s is the same as the $Re(Z_{\mathrm{film}})$ when the ω is infinite. The $Re(Z_{\mathrm{film}})$ at $\omega = \infty$ was calculated from the least-square fitting of the measured data over the frequency range from 1 to 6 GHz as shown in Fig. 6. Therefore, the tan δ of the $BaTi_4O_9$ thin film (tan δ_d) can be calculated from the Eq. (3).

The ε_r of $BaTi_4O_9$ thin films with various thicknesses and measured at 1–6 GHz were shown in Fig. 7. The ε_r of all the films is constant over the measured frequency range. The ε_r of the $BaTi_4O_9$ film with the thickness of 150 nm is approximately 36 and it slightly increased with the increase of the film thickness. For the $BaTi_4O_9$ film with the thickness of 460 nm, it was approximately 38. The ε_r of the bulk $BaTi_4O_9$ measured at 4 GHz was reported to be 39.[13] Therefore, the ε_r of the $BaTi_4O_9$ film is very similar to that of the bulk $BaTi_4O_9$. Fig. 7 also shows the tan δ of $BaTi_4O_9$ film (tan δ_d) and it is low, less than 0.05, especially for the film with the thickness of 460 nm, it was about 0.0001 at 1–3 GHz.

4. Conclusions

The dielectric properties of $BaTi_4O_9$ thin films were investigated over the microwave frequency range. From the measured impedance of the circular-patch capacitor, we successfully obtained the ε_r and the tan δ of the $BaTi_4O_9$ film up to 6 GHz.

The ε_r of the $BaTi_4O_9$ thin film slightly increased from 36 to 38 with the increase of the film thickness. These ε_r of $BaTi_4O_9$ thin films were similar to that of the $BaTi_4O_9$ ceramics, which is about 36–39. The tan δ of the $BaTi_4O_9$ film with 460 nm thickness was very low approximately, 0.0001 at 1–3 GHz. Therefore, the $BaTi_4O_9$ film is considered to have excellent dielectric properties and might be applicable for microwave thin film devices.

Acknowledgements

This work was supported by the Ministry of Science and Technology through the project of Nano-Technology and one of the authors also acknowledges that this work was financially supported by the Ministry of Science and Technology through NRL project.

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