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Microscopic observation of the temperature coefficient distribution of microwave materials using scanning electron-beam dielectric microscopy

Y. Cho*, A. Satoh, H. Odagawa

Research Institute of Electrical Communication, Tohoku University, 2-1-1 Katahira, Aoba-ku Sendai 980-8577, Japan

Abstract

Studies on scanning electron-beam dielectric microscopy (SEDM) are reported. This microscopy technique is used for determining the temperature coefficient distribution of dielectric materials using an electron-beam as a heat source. This microscopy technique, which has the ability to simultaneously observe SEM images and the material composition by EPMA, has a resolution better than that of photothermal dielectric microscopy. To demonstrate the usefulness of this technique, the two-dimensional image of a two-phase composite ceramic is measured. To shorten a measurement time, a new type of SEDM for measuring the real time transient response caused by a single pulsed electron-beam is also successfully developed.

Keywords: Capacitors; Dielectric properties; Dielectric temperature coefficient; Electron microscopy

1. Introduction

A new photothermal technique to evaluate the thermal properties of dielectric materials and to measure the microscopic distribution of the temperature coefficient of the dielectric constant has recently been developed using the photothermal dielectric (PTD) effect.[1,2] This method is based on the temperature characteristic of the dielectric constant of a light-irradiated material. Microscopic measurements of the distribution of the temperature coefficients of the dielectric constant provide more precise information for designing the material than that obtained from a macroscopic measurement.[3] In particular, in composite ceramics composed of grains with different dielectric temperature coefficients (for example, a positive and a negative coefficient), a microscopic technique to assess the distribution of the temperature coefficient of the dielectric constant is very effective for the precise characterization of the material.

This method of using a light beam as a heat source has the advantages of being relatively simple and inexpensive. However, its resolution is limited by the light wavelength and it is difficult to perform an in situ measurement identifying each grain of a ceramic and its composition. To give an answer to the above-mentioned problem, a new microscopy for determining the temperature coefficient distribution of dielectric constants was developed using an electron beam as a heat source instead of a light beam. In this paper, the results of the studies on scanning electron-beam dielectric microscopy is described, which has a resolution better than that of photothermal dielectric microscopy and with the ability of in situ observation of SEM images and of material compositions by electron probe micro analyzer (EPMA). To demonstrate the usefulness of this technique, we measured the two-dimensional image of a two-phase composite ceramic composed of TiO_2 and $Bi_2Ti_4O_{11}$ that have a positive and a negative dielectric temperature coefficient, respectively.

Finally, to shorten a measurement time, a new type of SEDM for measuring the real time transient response caused by a single pulsed electron-beam is also successfully developed.

2. Electron-beam dielectric microscopy with continuously chopped e-beam

A schematic diagram of the scanning electron-beam dielectric microscope (SEDM) system is shown in Fig. 1. To measure the capacitance variation with the chopped

* Corresponding author. Tel./fax: +81-22-217-5529.
E-mail address: cho@riec.tohoku.ac.jp (Y. Cho).

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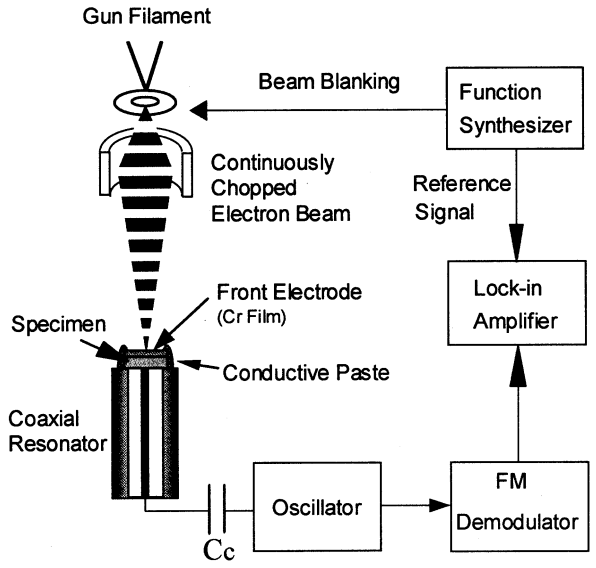

Fig. 1. Schematic diagram of the scanning electron-beam dielectric microscope system using continuously chopped e-beam.

electron-beam with an angular frequency ω_c, we used a coaxial cavity resonator with a capacitor made of the material to be measured. This probe was connected to an oscillator tuned to the resonance frequency of the probe. The alternating temperature change due to the absorption of the chopped electron-beam (e-beam) causes an alternating variation of the capacitance because of its temperature characteristic, so that the oscillation frequency is modulated by the change of the capacitance. As a result, a frequency modulation (FM) signal comes from this oscillator. By detecting this FM signal using the FM demodulator and a lock-in amplifier, we can obtain a voltage signal which is proportional to the capacitance variation.

In the one-dimensional analysis, the alternating variation in capacitance C_{ac} is given by[2]

$$C_{ac} = C(T_0)\frac{\varepsilon'(T_0)}{\varepsilon(T_0)}\frac{I_0}{2\ell\rho_s C_s}\frac{1}{\omega_c}\cos\left(\omega_c t - \frac{\pi}{2}\right) \quad (1)$$

for thermally thick material ($\mu_s \ll \ell$), and

$$C_{ac} = C(T_0)\frac{\varepsilon'(T_0)}{\varepsilon(T_0)}\frac{I_0}{2\sqrt{k_b\rho_b C_b}}\frac{1}{\sqrt{\omega_c}}\cos\left(\omega_c t - \frac{\pi}{4}\right) \quad (2)$$

for thermally thin material ($\mu_s \gg \ell$), where $\varepsilon(T_0)$ (F/m), $\varepsilon'(T_0)$ (F/m-k) and $C(T_0)$ (F) are the dielectric constant, its first-order temperature coefficient and the capacitance at room temperature T_0 (K), respectively. The terms ρ_s (kg/m^3), C_s (J/kg-K) and μ_s (m) indicate the density, the specific heat and the thermal diffusion length of the specimen, respectively. The terms k_b (W/m-K), ρ_b (kg/m^3) and C_b (J/kg-K) are the thermal conductivity, the density and the specific heat of the backing material, respectively. The terms ℓ (m), $\omega_c = 2\pi f_c$ (rad/s) and I_0 (W/m^2) indicate the thickness of the specimen, the chopping angular frequency of the incident electron-beam and the electron-beam intensity, respectively. The localized alternating temperature change due to the absorption of the focused chopped electron-beam causes a localized alternating variation in the capacitance because of its temperature characteristics. Thus, we obtain a two-dimensional image of the distribution of the dielectric temperature coefficient of dielectric constant by scanning the focused electron-beam with the SEM image and the EPMA image, simultaneously.

Using the above-mentioned SEDM system, some experiments were performed to reveal the fundamental characteristics of the microscopy.

A two-dimensional image of the dielectric temperature coefficient distribution of the two phase composite ceramic composed of TiO_2 and $Bi_2Ti_4O_{11}$[1,4] was observed with simultaneous measurements of the SEM image and EPMA. The SEM image and EPMA image are shown in Fig. 2 (a) and (b), respectively. The grains appearing black are composed of only TiO_2 and those appearing white are composed of only $Bi_2Ti_4O_{11}$. The EPMA image shows the X-ray emission strength of the Bi–M_α line. Therefore, the white dots in the EPMA image show the distribution of Bi atoms. As previously reported[1,4], TiO_2 has a large negative temperature coefficient of dielectric constant of $\tau_\varepsilon = -879$ ppm/°C, whereas $Bi_2Ti_4O_{11}$ has a large positive coefficient of $\tau_\varepsilon = 1034$ ppm°C. Thus the macroscopic average value of the temperature coefficients for TiO_2–$Bi_2Ti_4O_{11}$ ceramic is $\tau_\varepsilon = -34$ ppm°C, because of the cancellation of the coefficients which have opposite signs.

In this study, a probe with a resonant frequency of 940 MHz without the sample and of 837 MHz with the sample was used. The thickness of the polished sample was 50 μm. As the front electrode of the sample capacitor, we used a Cr film with a thickness of 300 Å.

Fig. 3 (a)–(c) shows the measured two-dimensional SEDM images of the temperature coefficient of dielectric constant taken from the same location as the SEM and EPMA images. These SEDM images were obtained with a beam current $I_p = 150$ nA, a beam accelerating voltage $V_a = 25$ kV and a beam chopping frequency $f_c = 100$ kHz [Fig. 3(a)], 10 kHz [Fig. 3(b)] and 1 kHz [Fig. 3(c)]. In order to obtain the only information that was purely dependent on the temperature coefficient of dielectric constant, the data at $f_c = 10$ kHz and 1 kHz were divided by a normalization factor of 10 and 100, respectively, because the absolute value of the PTD signal increases linearly with the reciprocal of the beam chopping frequency for thermally thick material [see Eq. (1)]. A clear temperature coefficient image corresponding to the different grain types was obtained only at a beam chopping frequency of $f_c = 100$ kHz, and the image became ambiguous and homogeneous as the beam chopping frequency was decreased. This can be

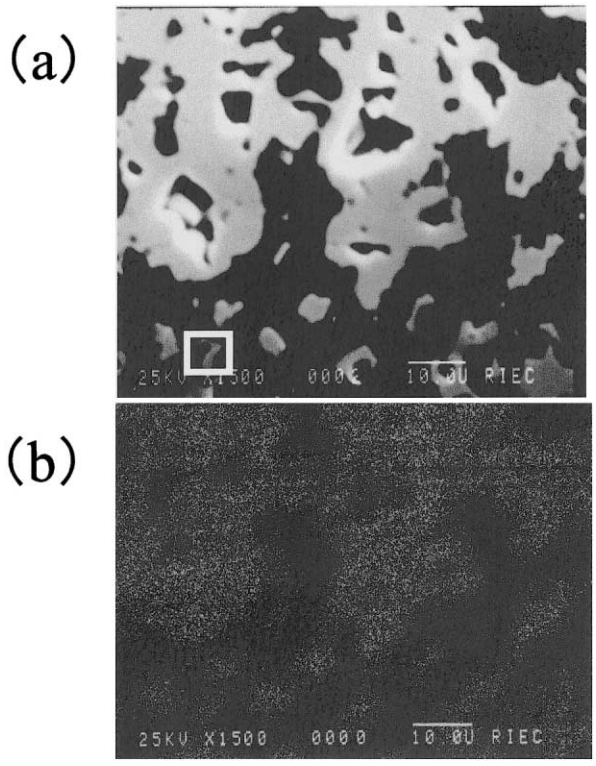

Fig. 2. SEM image (a) and EPMA (Bi–M_α line) image (b) of the grains of a TiO_2–$Bi_2Ti_4O_{11}$ ceramic sample.

explained qualitatively as follows. As is well known, the thermal diffusion length μ_s of the material is inversely proportional to the square root of the beam chopping frequency f_c.[5] Therefore, we can obtain a well resolved image corresponding to the grains with opposite signs of temperature coefficient when we use a high chopping frequency of 100 kHz. As the beam chopping frequency decreases, μ_s becomes longer and a hemisphere with a radius of μ_s from the sample surface is alternately heated simultaneously. Thus, we obtained a homogeneous and averaged temperature coefficient image corresponding to the macroscopic value of the temperature coefficient of $\tau_\varepsilon = -34$ ppm/°C of the two-phase TiO_2–$Bi_2Ti_4O_{11}$ at a low f_c (1 kHz). The well resolved images Fig. 3(a) and Fig. 2 show a good correlation between the temperature coefficient distribution image and the ceramic grains. This SEDM method is very useful for a simultaneous measurement of temperature coefficient of dielectric constant identifying each grain of a ceramic and its composition.

Next, the very small area of the specimen, which is surrounded by the white square in Fig. 2(a), was measured. The magnified SEDM image are shown in Fig. 4, respectively. In this case, a beam chopping frequency was $f_c = 100$ kHz. Although, a highly resolved SEDM image well correlated with the SEM image was obtained, there is no positive area in the image. That is, we failed to determine the sign of the temperature coefficient of the dielectric constant of the very small $Bi_2Ti_4O_{11}$ grain (the white area in the SEM image). This

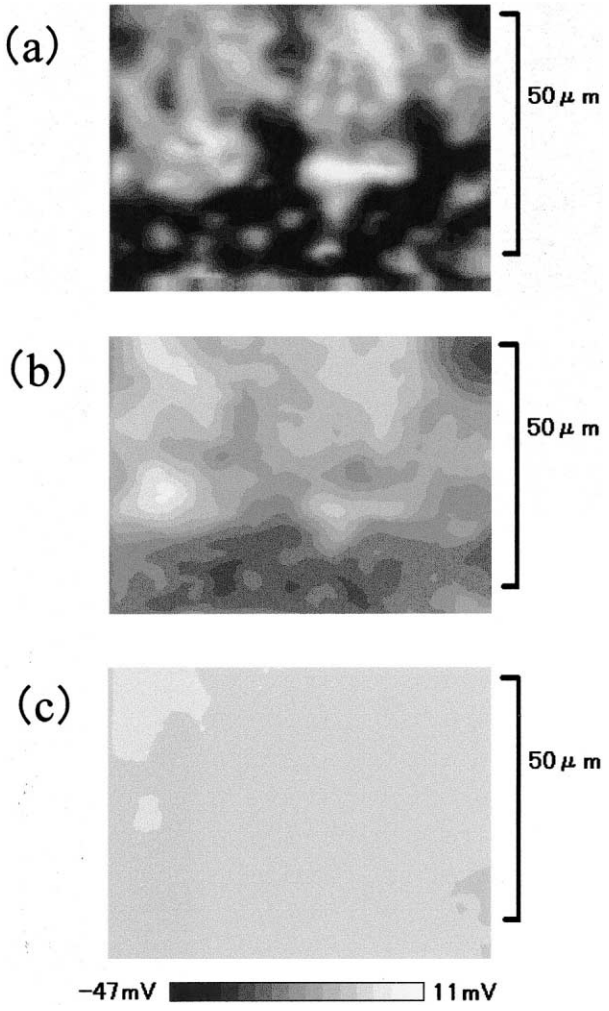

Fig. 3. Dielectric temperature coefficient images of TiO_2–$Bi_2Ti_4O_{11}$ ceramic by SEDM, using a beam-chopping frequency of (a) 100, (b) 10, and (c) 1 kHz. These images were taken from the same location as those of Fig. 2.

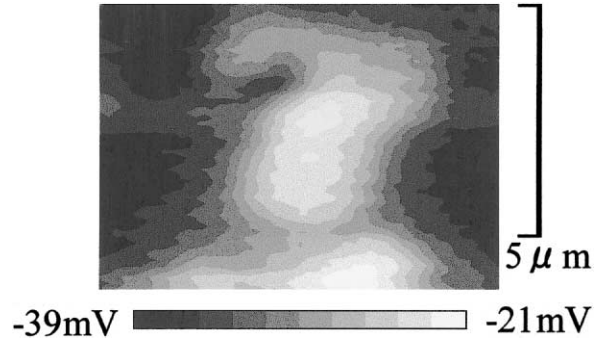

Fig. 4. Magnified SEDM image of the area enclosed within the square shown in Fig. 2.

result indicates that even at $f_c = 100$ kHz (which is the limit of our present equipment), the thermal diffusion length μ_s is too long to resolve (or determine) the temperature coefficient distribution of the small area precisely. To resolve this small $Bi_2Ti_4O_{11}$, grain correctly, we will

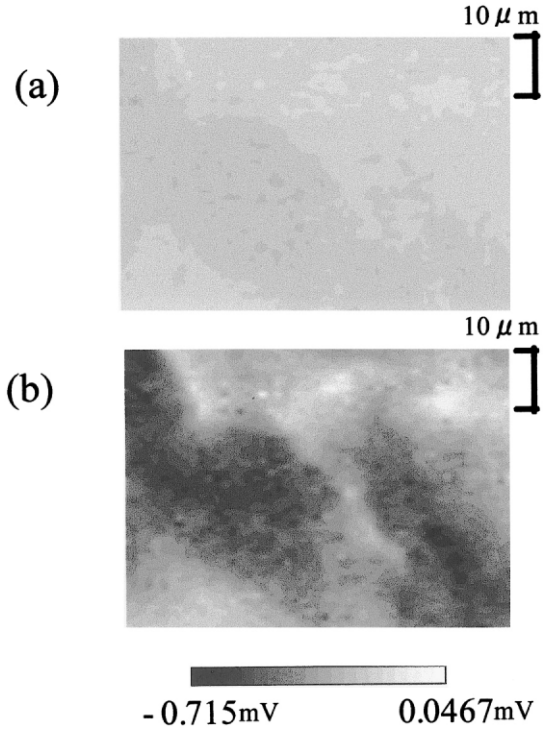

Fig. 5. SEDM images taken by using a single pulsed e-beam of TiO_2–$Bi_2Ti_4O_{11}$ ceramic. The data were analyzed into equivalent SEDM images with each chopping frequency of 0.4 (a) and 6 kHz (b) by using FFT technique.

need to use a thermal wave with a very short, i.e. submicron, wavelength (thermal diffusion length). We also need a higher beam chopping frequency to obtain a submicron resolution.

The above mentioned SEDM imaging technique using a continuously chopped e-beam is very accurate for measuring a dielectric temperature coefficient distribution. However, to obtain the several SEDM images as a function of beam chopping frequency, it requires a long measurement time. To avoid the data drifts caused by the variation of the measurement circumstances such as a room temperature drift, it is very important to shorten the measurement time. Therefore, we developed a new SEDM system for the measurement of the electron-beam thermal dielectric response caused by a single pulsed e-beam. In the new system, we used a digital oscilloscope for a broad-band detection of the pulsed signal instead of a lock-in amplifier in Fig. 1.

After we confirmed that we were able to detect a transient response to the single pulsed incident e-beam with enough dynamic range and sensitivity, we performed a two-dimensional scanning on the same specimen using a focused e-beam pulse. The results are shown in Fig. 5 with the SEM image of the same location. These images were produced in accordance with the following procedure.

At first, we accumulated the pulse response data only in one time scanning, and then these data were analyzed into several numerical data which correspond to each image with arbitrary beam chopping frequency using fast Fourier transformation (FFT) calculation. As a result, we can synthesize the equivalent images to those taken by an SEDM using a continuously chopped e-beam with an arbitrary beam chopping frequency. From the Fig. 5, it is understood that we can obtain the SEDM image with arbitrary beam chopping frequency only by one time scanning. Thus, we succeeded to make a remarkable reduction of the measurement time.

3. Conclusion

A new technique to assess the distribution of the temperature coefficient of dielectric materials for microwave applications has been proposed. This microscopy technique is very useful for determining the relative value of the temperature coefficient of the dielectric constant of each grain in a composite ceramic with a high resolution and with the ability of in situ SEM and EPMA observations giving grain shapes and material compositions.

Finally, to reduce the measurement time of SEDM images with several e-beam chopping frequency, we succeeded to develop a new SEDM which detects the transient response of an electron beam thermal dielectric signal caused by a pulsed e-beam.

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