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Dielectric properties and relaxation of $SrBi_2(Nb_{0.25}Ta_{0.75})_2O_9$ ceramic at RF frequency

Jun Zheng Liu*, K.W. Kwok, H.L.W. Chan, C.L. Choy

Department of Applied Physics and Materials Research Centre, The Hong Kong Polytechnic University, Hung Hom, Hong Kong, China

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Abstract

The dielectric properties and relaxation of the $SrBi_2(Nb_{0.25}Ta_{0.75})_2O_9$ ceramic were investigated from 3 to 300 MHz using frequency domain measurement by an impedance spectroscopy. Our results show that the dielectric relaxation of the ceramic can be understood in terms of power law dependence known as the Curie–von Schweidler law. It is assumed that defects, Schottkey jumps of electrons between localized states and the motions of ions play an important role in the dielectric properties and relaxation of the ceramic. The ceramic has large relative permittivity ($\varepsilon_r \sim 117$ at 300 MHz), low dielectric loss ($\tan\delta_e < 0.018$ at 300 MHz) and small dispersion ($<3\%$ decrease in ε_r from 3 to 300 MHz). The Curie temperature of the ceramic is very high (~ 370 °C), implying that it has a small temperature coefficient. Hence the $SrBi_2(Nb_{0.25}Ta_{0.75})_2O_9$ ceramic is a potential candidate for microwave applications.

Keywords: Dielectric properties; Microwave applications; Relaxation; RF frequency; $SrBi_2(Nb_{0.25}Ta_{0.75})_2O_9$ ceramic

1. Introduction

Along with the rapid developments of wireless telecommunication systems operating in UHF band and high speed computing with CPU frequency higher than 1 GHz, there have been growing interest in investigating microwave properties of ceramic materials and increasing demand for new ceramic materials with better microwave properties.[1–3] In general, ceramics with large relative permittivity, low dielectric loss and small temperature coefficient are required for microwave device applications. The larger the relative permittivity, the smaller the microwave device components can be. A low dielectric loss can improve the insertion loss and make higher operating frequency possible. A number of ceramic materials, e.g. TiO_2, Al_2O_3, $Ba_2Ti_9O_{20}$ and $(Zr_xSn_{1-x})TiO_4$, have been extensively studied for microwave applications. However, each of the ceramics has drawbacks. TiO_2 has large relative permittivity and low dielectric loss, but its temperature coefficient is very large ($\sim$450 ppm/K), making the ceramic not suitable for practical use. In contrast, $Ba_2Ti_9O_{20}$ and $(Zr_{0.8}Sn_{0.2})TiO_4$ have very low temperature coefficient, but their relative permittivities are not very high ($\sim$40), hence limiting their use in miniaturized microwave devices.

The layer-structured bismuth compounds is one of the most technologically interesting materials and has been extensively studied since 1960s.[4–6] It has the general formula: $(Bi_2O_2)^{2+}(A_{m\text{-}1}B_mO_{3m+1})^{2-}$, where A is usually a divalent ion, such as Sr, Ba or Pb, and B is Ti^{4+}, Nb^{5+} or Ta^{5+}. m and $m-1$ are the numbers of oxygen octahedra and pseudo-perovskite units, respectively, in the pseudo-perovskite layer $(A_{m-1}B_mO_{3m+1})^{2-}$, which is separated at intervals by the bismuth oxide layer $(Bi_2O_2)^{2+}$. It has been shown that $SrBi_2Ta_2O_9$ (SBT) and $SrBi_2Nb_2O_9$ (SBN) ceramics have a very high phase transition temperature ($\sim$420 °C), and hence their relative permittivity has a weak temperature dependence at room temperature. In other words, their temperature coefficient is small and should be applicable to microwave device applications. Our previous work[7] has shown that the $SrBi_2(Nb_{0.25}Ta_{0.75})_2O_9$ ceramic has a large relative permittivity ($\sim$120) and low dielectric loss ($\sim$0.02) in the frequency range of 100 Hz to 100 kHz at room temperature, indicating that the ceramic is a potential

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* Corresponding author. Tel.: +852-27667797; fax: +852-23337629.

E-mail address: apliujz@polyu.edu.hk (J.Z. Liu).

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candidate for microwave devices applications. In the present work, we further investigate the dielectric properties of the $SrBi_2(Nb_{0.25}Ta_{0.75})_2O_9$ ceramic at RF frequency range of 3 to 300 MHz.

2. Experimental

In the present work, the $SrBi_2(Nb_{0.25}Ta_{0.75})_2O_9$ (SBNT) ceramic was prepared by the conventional mixed-oxide method. The raw materials used, $SrCO_3$, Bi_2O_3, Ta_2O_5 and Nb_2O_5, are all of chemically pure grade. The above raw materials in the stoichiometric ratio of the composition were first mixed thoroughly by ball milling. After that, the material was calcined at 850 °C for 8 hs to form the $SrBi_2(Nb_{0.25}Ta_{0.75})_2O_9$ composition. The calcined compound was ball-milled again to reduce the particle size, and then mixed with a binder and pressed into disc samples of diameter 16 mm and thickness 0.4 mm. After removal of the binder, the disc samples were sintered in sealed Al_2O_3 crucibles at 900 °C for 1 h and then at 1065 °C for 45 min for crystallization. The crystallite structure of the sintered sample was investigated using an X-ray diffractometer (XRD) with nickel-filtered Cu K_α radiation (X'pert System, Philips Electronic Instruments), and the microstructure was examined using a scanning electron microscope (Leica Stereoscan 440).

Silver electrodes were applied on the top and bottom surfaces of the sample by screen printing and firing at 600 °C for the subsequent electrical measurements. A LCR meter (SR720 Stanford Research Systems) was used to measure the relative permittivity ε_r at 1 and 100 kHz as a function of temperature for determining the Curie transition temperature of the sample. The ε_r value in the frequency range of 300 MHz was measured using a RF impedance analyzer (HP 4291B) at room temperature.

3. Results and discussion

The X-ray diffraction pattern of the SBNT ceramic is shown in Fig. 1. It can be seen that the sample exhibits a well-crystallized phase with peaks attributable to the layered SBNT, and possesses a nontextured polycrystalline structure with no evidence of secondary phase. Fig. 2 shows the SEM micrograph of the fracture surface of the SBNT sample. It is seen that the sample consists of anisometric plate-like crystalline grains of size in the range of 2–4 μm. A few holes are observed in the sample, the observed density of the sample is about 7100 kg/m^3, which is 87% of the theoretical density.

The variation of ε_r at 1 and 100 kHz with temperature in the range of −170 to 430 °C is shown in Fig. 3. The observed ε_r values at 1 and 100 kHz are almost the same in the measurement temperature range. They have a value of about 120 at 25 °C, and increases slowly as temperature increases to 130 °C. At higher temperatures, the observed ε_r value increases much faster with temperature and exhibits a phase transition with a maximum value of about 520 at 370 °C. Above the temperature, the ε_r value can be approximately described by the Curie–Weiss law $\varepsilon = C/(T-\theta)$. The corresponding Curie–Weiss temperature θ is 311 °C and the Curie constant C is 3.12×10^5 °C, which has the same order of magnitude as those of other ferroelectric oxides such as $BaTiO_3$, $Cd_2Nb_2O_7$ and $PbNb_2O_6$. As shown in Fig. 3, the phase transition temperature is almost

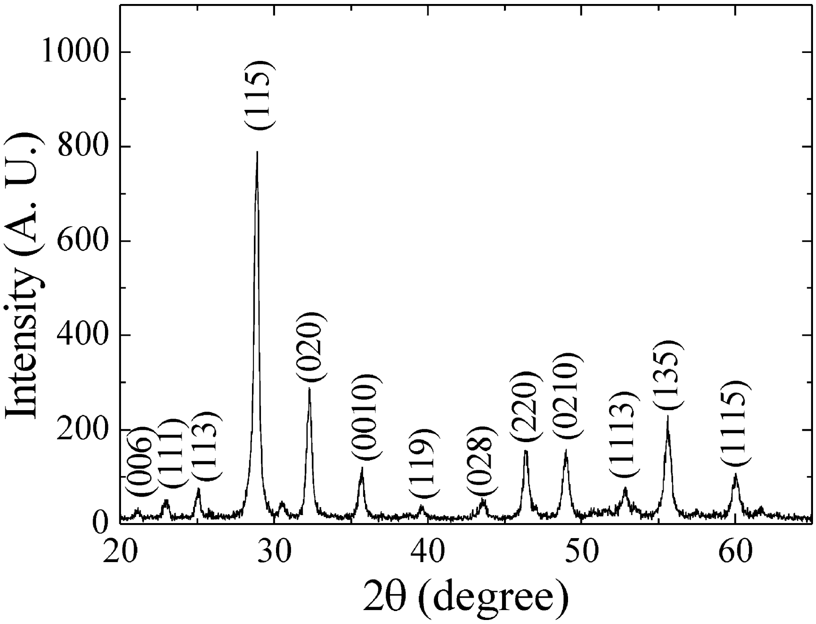

Fig. 1. XRD pattern of the $SrBi_2(Nb_{0.25}Ta_{0.75})_2O_9$ ceramic.

unchanged and the maximum value of ε_r decreases slightly with increasing frequency. These indicate that the SBNT ceramic is not a relaxor-type material.

The room temperature relative permittivity of the ceramic is shown as a function of frequency in Fig. 4. At 3 MHz, the observed ε_r value is about 120, and decreases slightly, by about 3%, as frequency increases to 300 MHz. On the other hand, the dielectric loss ($\tan\delta_e$) varies from 0.020 to 0.018 in the measurement frequency range. The small dielectric dispersion can be considered as a series of dielectric relaxations continuously distributed throughout the frequency range. It is also noted that the observed ε_r value follows a power law dependence on frequency (f), which is known as the Curie–von Schweidler law:

$$\varepsilon_r = a + bf^{n-1} \quad (1)$$

where the power n ($n < 1$) is the dispersion parameter, a

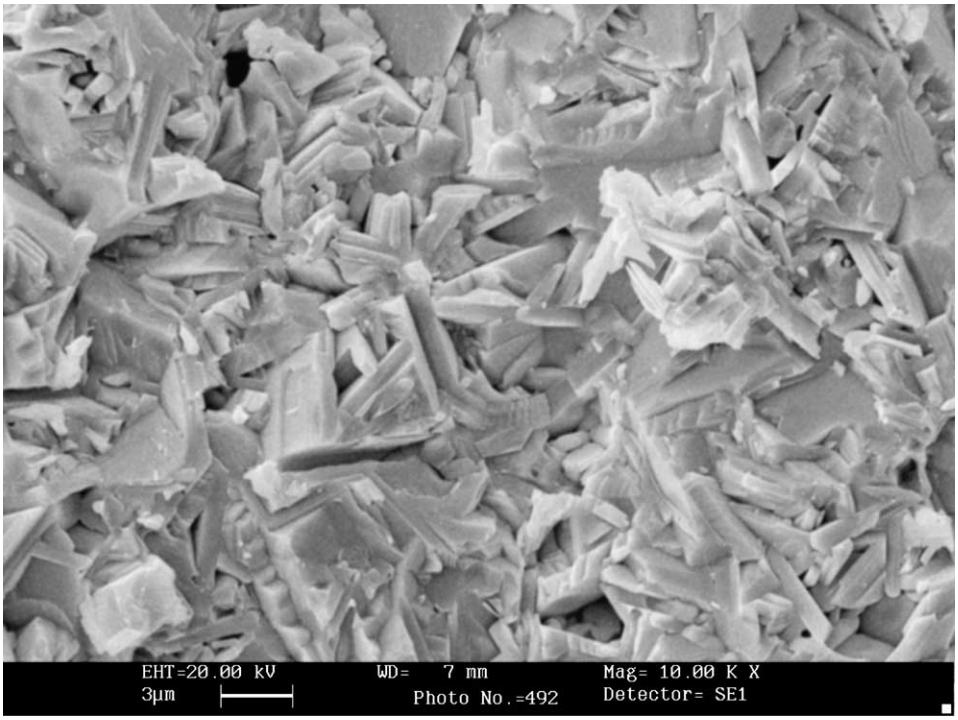

Fig. 2. SEM micrograph of the fracture surface of the $SrBi_2(Nb_{0.25}Ta_{0.75})_2O_9$ ceramic.

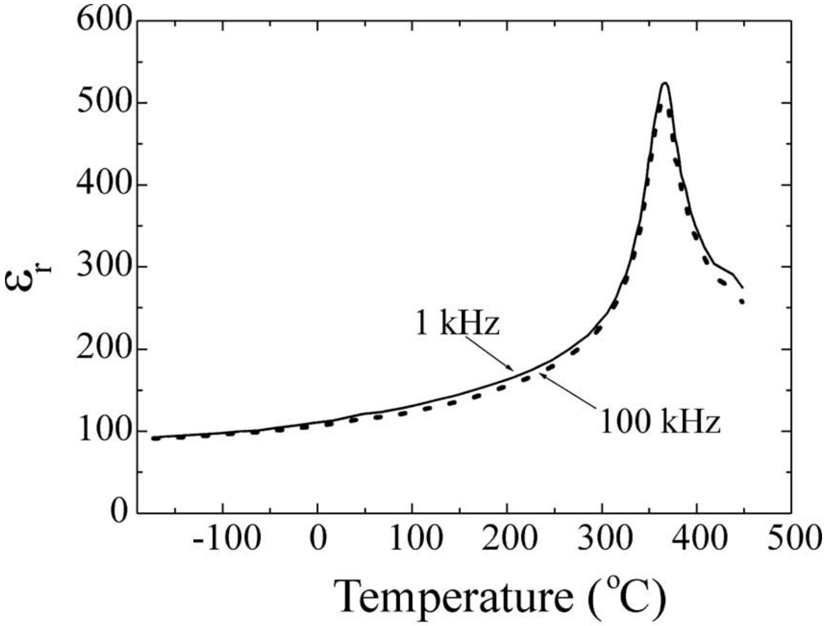

Fig. 3. Relative permittivity ε_r at 1 and 100 kHz of the $SrBi_2(Nb_{0.25}Ta_{0.75})_2O_9$ ceramic as a function of temperature.

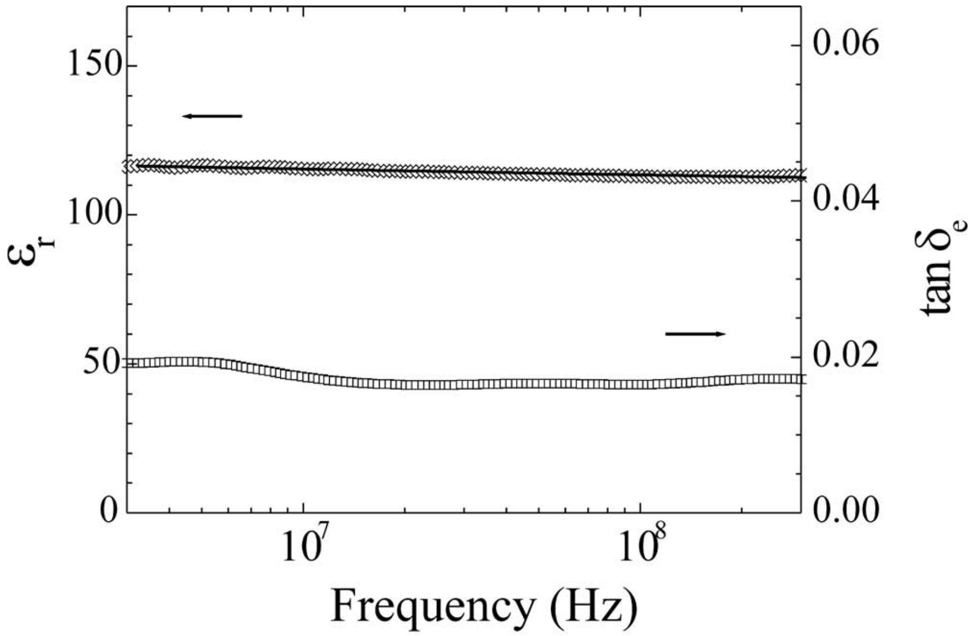

Fig. 4. Relative permittivity ε_r (◇) and loss factor tanδ_e (□) of the $SrBi_2(Nb_{0.25}Ta_{0.75})_2O_9$ ceramic as functions of frequency at room temperature. Solid line is fitted curve.

and b are temperature-dependent parameters. By fitting the experimental data to Eq. (1), the values of the parameters are determined ($n=0.97$, $a=84.7$ and $b=48.6$), and the calculated ε_r values are shown as a solid line in Fig. 4. The good agreement between the observed and calculated ε_r values implies that due to the dispersive nature of the material, there are relaxation currents which also follow a power law time dependence of approximately $\propto t^{-n}$.[8,9] Because of the t^{-n} behavior, the relaxation currents dominate at a short time interval, but the true leakage current exists for longer time. This is in agreement with the large dispersion parameter ($n \sim 0.97$), which means a large distribution of relaxation time in the ceramic.

It is recognized that intrinsic losses are associated with phonon scattering processes. Because of the layered structure of the SBNT ceramic, it is difficult to have very dense microstructure, so the intrinsic loss cannot be very low. Moreover, point defects, especially oxygen defects, are known to have defect-wells associated with and/or trapped electrons to the defect-wells and exhibit an effective charge. The imposed RF field excites these defects. In other words, the RF field is believed to generate acoustic oscillations and phonon emission at these defects thus resulting in extrinsic defect loss[10] with a short responding time.

Besides, thermally stimulated Schottky jumps of localized electrons with a medium time and the motion of ions with a long time contribute to the relative permittivity and dielectric loss.[9] However, the ions relax in a long time and contribute with an approximately constant term in the frequency range of our measurement. Therefore, the parameter a which is the frequency-independent part of the observed permittivity consists of the contribution of both localized electrons and ions.

4. Conclusion

The $SrBi_2(Nb_{0.25}Ta_{0.75})_2O_9$ (SBNT) ceramic has been prepared by the conventional mixed-oxide method. The ceramic exhibits a well-crystallized phase and possesses a nontextured polycrystalline structure. Dense microstructure has also been observed using the scanning electron microscope. The ceramic has relatively large relative permittivity ($\varepsilon_r \sim 117$ at 300 MHz), low dielectric loss (tanδ_e <0.018 at 300 MHz) and small dispersion ($<3\%$ decrease in ε_r from 3 to 300 MHz). The dielectric relaxation of the ceramic can be understood in terms of the Curie–von Schweidler relaxation law from 3 to 300 MHz. Defects, Schottky jumps of electrons between localized states and the motions of ions play an important role in the dielectric properties and relaxation of the ceramic.

Acknowledgements

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