On the measurement methods for dielectric constant determination in Nb/BaTiO$_3$ ceramics

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Abstract—**In this paper the measurement methods for dielectric constant determination in Nb/BaTiO$_3$ ceramics have been presented. The experimental results obtained using LCR meter with dielectric test fixture and the influence of ceramic microstructure on the accuracy of methods have been discussed.**

Keywords-ceramics; dielectric constant; measurement methods

I. INTRODUCTION

For accurate measurement of the dielectric constant of a ceramics, a three-terminal system is needed [1]-[4]. The parallel plate measurement methods, also called the three-terminal methods in ASTM D150 [5], involve the use of a parallel plate capacitor as a holder, with sample of the material under the test (MUT) sandwiched between the plates. These methods require an impedance analyzer or LCR meter. The measurements are at low frequencies. Typically, the methods use a wide frequency range from 20 Hz to 1 GHz. The MUT is biased by an AC source and the actual voltage across the material is monitored. The material test parameters, such as dielectric constant, are calculated by knowing the dimensions of the MUT and by measuring its capacitance.

This paper presents experimental results of capacitance measurement and dielectric constant determination of Nb/BaTiO$_3$ ceramics over a wide frequency range. The results are obtained with two methods: contacting electrode and non-contacting electrode method. The dielectric constant values in function of frequency are considered. The influence of samples microstructure on the accuracy of the methods is also discussed in details.

II. PARALLEL PLATE MEASUREMENT METHODS

There are two types of parallel plate measurement methods: contacting electrode method, and non-contacting electrode method. The least accurate measurements are by contacting electrode method without thin film electrode. When thin film electrode is applied onto surfaces of the MUT, the highest accuracy is possible. Therefore, contacting method is the most widely used one [6]-[7]. The main advantage of contacting method is that for obtaining the dielectric constant it is necessary to take only one measurement. On the other hand, the non-contacting electrode method has medium measurement accuracy and involves very simple preparation of sample and setup. MUT should be solid material with a flat and smooth surfaces, but dielectric constant is derived by using the results of two capacitance measurements.

When measuring the dielectric material between two electrodes, stray capacitance or edge capacitance is formed on the edges of the electrodes and consequently the measured capacitance is larger than the capacitance of the MUT. The edge capacitance causes a measurement error, since the current flows through the dielectric material and edge capacitor [8]-[9]. A solution to the measurement error is to use the guard electrode which absorbs the electric field at the edge and the capacitance that is measured between the electrodes is only composed of the current that flows through the dielectric material (Fig. 1). When the main electrode is used with a guard electrode, the main electrode is called the guarded electrode.

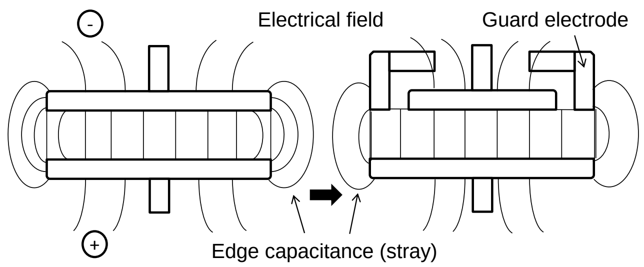

Figure 1. Illustration of guard electrode effect

A. Contacting electrode method

The contacting electrode method determines permittivity by measuring the capacitance of the MUT directly (Fig. 2). The dielectric constant is calculated using the equation:

$$\varepsilon_r = \frac{t \cdot C_p}{S \cdot \varepsilon_0} = \frac{t \cdot C_p}{\pi \left(\frac{d}{2}\right)^2 \cdot \varepsilon_0}, \tag{1}$$

where t is average thickness of MUT, C_p is equivalent parallel capacitance of MUT, S is guarded electrode's surface area, d is diameter, ε_0 is permittivity of vacuum. The contacting electrode method doesn't require material preparation and the operation involved when measuring is simple [7]. When contacting the MUT directly with the electrodes, an airgap is formed between the MUT and the electrodes. Materials with rough surfaces can be affected by airgap, as illustrated in Fig. 3. A measurement error can occur because the measured capacitance will be the series connection of the capacitance of the dielectric material

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and the airgap. Thin samples are most prone to airgap effect. The methods for reducing error due to airgap are formation of thin film electrodes on a dielectric material and maximization of the pressure on the test fixture to the extent that it doesn't deform the MUT. There is a technique to apply a thin film electrode onto the surfaces of the dielectric material in order to increase the contacting area and to eliminate the airgap that occurs between the MUT and the electrodes. An electrode is manufactured onto the dielectric material using high-conductivity silver paste. Thin film electrode must be thinner than the dielectric material. In this case, it is important to appropriately position the fabricated thin film electrode onto the MUT, to precisely contact the electrodes of test fixture. The measurement error is a function of the relative permittivity (ε_r) of the MUT, thickness of the MUT (t_m), and the airgap's thickness (t_a), and it is determined by:

$$\delta_{r\varepsilon} = \frac{\varepsilon_r - 1}{\varepsilon_r + \frac{t_m}{t_a}} \cdot 100\ [\%]. \tag{2}$$

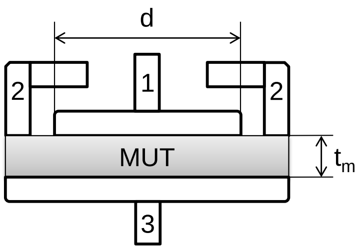

Figure 2. Illustration of contacting electrode method (1 – guarded electrode, 2 – guard electrode, 3 – unguarded electrode)

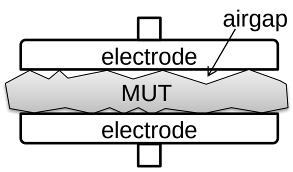

Figure 3. Illustration of rough-surfaced MUT

B. *Non-contacting electrode method*

The non-contacting electrode method does not require thin film electrodes, but still overcomes the airgap effect, so this method was conceptualized to unite the advantages and eliminate the disadvantages of the contacting electrode method [7]. Two capacitance measurements, obtained with the MUT and without it, are necessary and the results are used to derive dielectric constant as:

$$\varepsilon_r = \frac{1}{1 - \left(1 - \frac{C_1}{C_2}\right) \cdot \frac{t_g}{t_m}}, \tag{3}$$

where C_1 is capacitance without MUT inserted, C_2 is capacitance with MUT inserted, t_g is electrode gap, and t_m is average thickness of MUT. The gap between guarded/guard electrode and MUT (t_g-t_m) should be very small when compared to the thickness of the MUT (t_m) (Fig. 4).

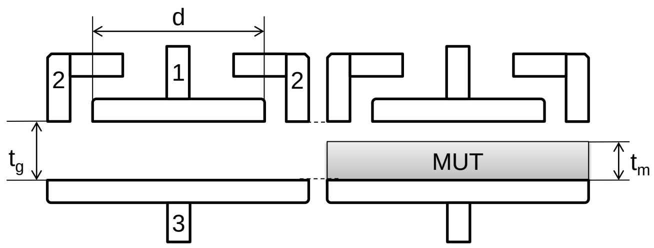

Figure 4. Illustration of non-contacting electrode method (1 – guarded electrode, 2 – guard electrode, 3 – unguarded electrode)

III. Sample Preparation and Experimental Procedure

The samples of modified $BaTiO_3$ ceramics doped with 0.5, 1.0, 2.0 mol % Nb_2O_5 were examined. The samples were prepared by a conventional solid state sintering procedure starting from reagent grade Nb_2O_5 and $BaTiO_3$ powder. Starting powders were ball milled in ethyl alcohol for 24 h. After drying at 200°C for several hours, the powders were pressed into disks under 120 MPa. The pellets were sintered at 1320°C in air atmosphere for 2 h [10]. The microstructure of the sintered samples were examined by scanning electron microscope (SEM) JOEL-JSM 5300 equipped with EDS (QX 2000S) system. Thin samples of 11 mm diameter and nearly 1.7 mm thickness were sandwiched using uniformly coated silver paste on both sides so that a ceramic capacitor structure is formed. The calculation of dielectric constant (permittivity) was performed by using measured values of capacitance, samples thickness and electrode area. The dielectric properties of the samples are measured in the frequency range from 20 Hz to 1 MHz.

The measurement system that employ the parallel plate methods consists of LCR meter Agilent 4284A [11] with the 16451B dielectric test fixture [12], which has capabilities to measure solid materials. Four electrodes are implemented to accommodate the contacting and non-contacting electrode methods and various MUT sizes. The three-terminal configuration with guard electrode eliminates the effect of edge capacitance and prevents occurrence of larger capacitance value than it really is. First electrode is unguarded electrode which is fed to the measurement instrument's high terminal, and the low terminal is connected with guarded electrode. Guard electrode is connected to the outer conductor of the BNC connector and encompasses the main electrode so that absorbs the electric field at the edge of the electrodes. When 16451B test fixture is used, measurements are possible in the frequency range up to 30 MHz, with maximum DC voltage of ±42 V. This test fixture has wide operation temperature range (from 0°C to 55°C). The limitation is cable length (1m). Since the 16451B test fixture introduces errors due to electrical length, residual impedance, and stray admittance, these errors can be entirely removed by open, short and load compensation. Measurement procedure flow chart with test fixture is shown in Fig. 5. The MUT must be a solid sample that is smooth and has equal thickness from one end to the other end.

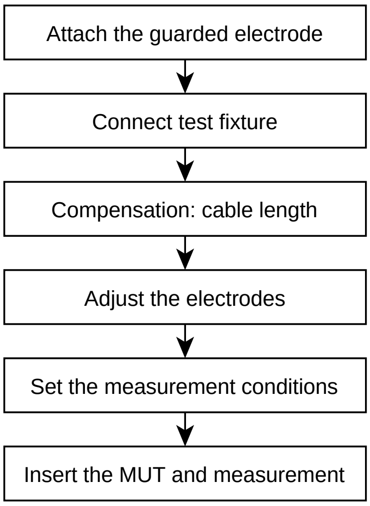

Figure 5. Basic flow chart for measurement with 16451B test fixture

IV. Results and Discussion

$Nb/BaTiO_3$ samples are stable and don't transform under applied pressure. Because of that, samples are suitable for both methods: contacting and non-contacting. The SEM imaging showed that the samples of Nb doped $BaTiO_3$ ceramics have spherical shaped grains (not flat surface). For 0.5 mol% of $Nb/BaTiO_3$ samples with fairly uniform microstructure, the average grain size is from 0.5 µm to 1 µm (Fig. 6). With increasing of dopant concentration, the grain size of $Nb/BaTiO_3$ samples increases and for the samples doped with 2 mol% of Nb the grain sized is around 7 µm, dispersed in a fine-grained matrix with average grain size in the range from 0.5 µm to 2 µm (Fig. 7). The Nb - rich regions are associated with small grained microstructure. Apart from the fine grains, some local areas have secondary abnormal grains of dopant [10], so the airgaps are deeper.

The dielectric constant evaluation has been made by capacitance measurements with two different methods in frequency range from 20 Hz to 1 MHz. According to the results plotted in Fig. 8, the permittivity for all samples decreases with increase of dopant concentration. At room temperature, the highest value of dielectric constant is ranged from 2600 for samples doped with 2 mol% to 3250 for the investigated samples doped with 0.5 mol% of Nb. The lowest value of dielectric constant (ε_r=2600) measured in samples doped with 2 mol% of Nb can be attributed to the formation of secondary abnormal grains that clearly lead to the decrease of permittivity. Moreover, the inhomogeneous distribution of dopand and formation Nb rich regions causes decreasing dielectric constant as well. After an insignificantly higher value of dielectric constant at low frequencies, ε_r becomes nearly constant at frequencies greater than 400 kHz.

As can be seen from the results show in Fig. 8, measured characteristics of $Nb/BaTiO_3$ samples with contacting and non-contacting electrode methods are quite consistent. It can be concluded that these methods can be equally used for dielectric characterization of $BaTiO_3$ ceramics. The relative deviation of dielectric constant between methods is shown in Fig. 9. The largest relative deviation was observed at low frequencies, where the samples have a maximum of dielectric constant. The deviation is in the range from 0.01 % to 3.2 %. Due to the changes of the instrument measurement range, it can be seen increase in relative deviation at 200 kHz.

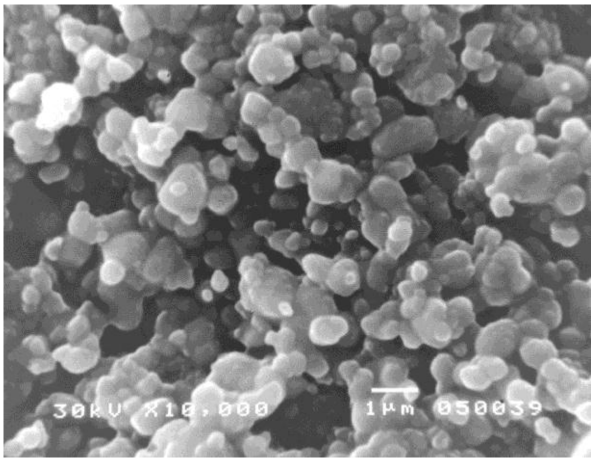

Figure 6. SEM image of 0.5 mol% $Nb/BaTiO_3$ sintered at 1320°C

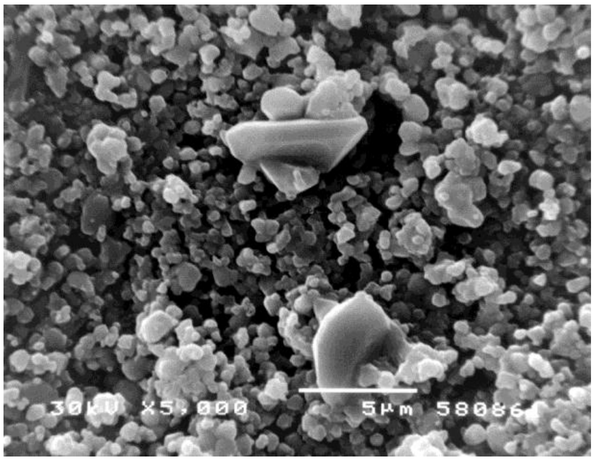

Figure 7. SEM image of 2 mol% $Nb/BaTiO_3$ sintered at 1320°C

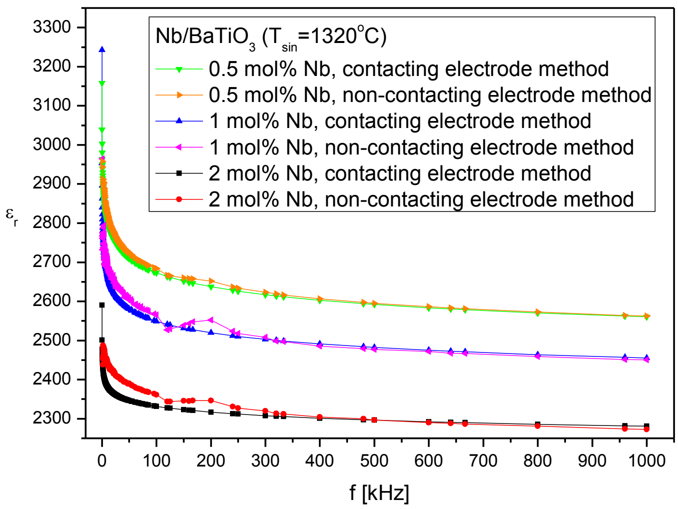

Figure 8. Dielectric constant as function of frequency for $Nb/BaTiO_3$ samples measured with contacting and non-contacting electrode methods

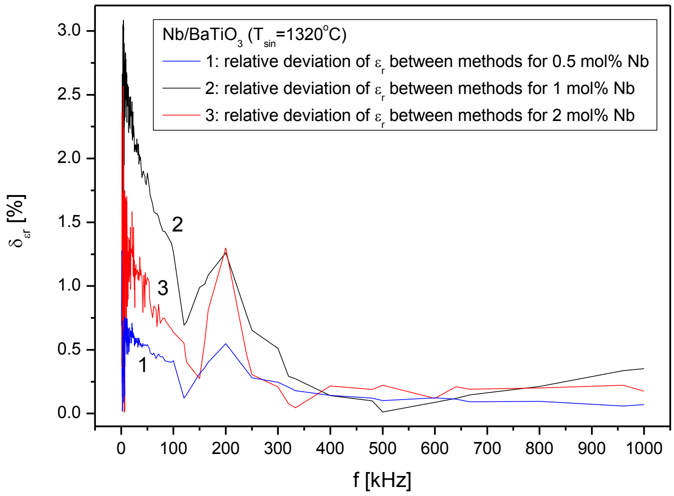

Figure 9. Relative deviation of dielectric constant between methods for $Nb/BaTiO_3$ samples

To achieve stable measurements with contacting electrode method, the pressure should be set at a level that doesn't deform the MUT. The pressure should be as strong as possible in order to minimize the occurrence of the airgap between the MUT and the electrodes. When the non-contacting method is employed, the electrode gap (t_g) is required to be at most 10% larger than the thickness of the MUT. The results of measurement error caused by airgap using (2) have been calculated in Table 1. The relative error is greater with increasing of t_a/t_m ratio and dielectric constant. Notice that the effect is higher with thin materials.

TABLE I. MEASUREMENT ERROR CAUSED BY AIRGAP

ε_r / t_a/t_m	2500	3000
0.001	71.4 %	74.98 %
0.005	92.56 %	93.72 %
0.01	96.12 %	96.74 %
0.05	99.17 %	99.30 %
0.1	99.56 %	99.63 %

V. CONCLUSION

In this paper the measurement methods for dielectric constant determination in $Nb/BaTiO_3$ ceramics have been investigated. The samples of modified barium titanate ceramics were used to compare contacting and non-contacting electrode methods. These three-terminal methods were used to eliminate a measurement error caused by the edge capacitance. The thin film electrodes were prepared by silver paste to cover the roughness on the samples surfaces and to reduce error due to airgap. Furthermore, it is noticeable that measurement error increases with increasing of airgap thickness. Very good results in a matching of $\varepsilon_r(f)$ plot for both methods are achieved. The maximum relative deviation between methods is 3.2%.

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