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The 11th EMAP Conference

Proceedings

December 1(Tue.) – 3(Thu.), 2009

The 11th International Conference on Electronic Materials and Packaging

Venue:
The Gurney Resort Hotel and Residences

Hosted by:
School of Mechanical Engineering, USM

Organizer

Co-organizer

Sponsors

EMAP 2009, Penang, Malaysia, EP09 27-11

EMAP 2009

Development of bismuth titanate for wireless dielectric antenna application

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Abstract—**Perovskite structure of bismuth titanate (is referred as undoped sample) and neodymium doped-bismuth titanate (is referred as doped sample) has been widely studied for nonvolatile random access memories (NvRAMs) because of its outstanding ferroelectricity. However, in our study these compounds were developed to evaluate the potential as dielectric antenna for wireless application. The raw materials i.e. bismuth nitrate penthahydrate, neodymium nitrate hexahydrate and titanium (IV) isopropoxide were used to synthesize those compounds via fuel-free combustion route. The as-synthesized powders were subsequently sent for the calcination at 800°C for 3hrs. An intermediate phase of $Bi_{12}TiO_{20}$ was observed in the undoped sample but it was not appeared in the doped samples. This result showed that the formation of such intermediate phase was reduced with neodymium addition. The microstructure of bismuth titanate was characterized by plate-like structure and its size was observed to decrease with increasing the neodymium addition. It showed that neodymium act as a grain growth inhibitor in bismuth titanate compound. In dielectric properties stand-point, the doped sample has higher the dielectric value as compared to undoped sample. Moreover, the dissipation factor of doped sample is lower than undoped sample by about 1 to 1.5 times.**

Keywords: **bismuth titanate, grain, dielectric properties**

I. INTRODUCTION

Bismuth titanate, $Bi_4Ti_3O_{12}$ is one of the widely used materials in electronics industry because of its high dielectric constant with particularly appropriate doping [1]. The substitution of lanthanides element in this compound has been used in many studies in order to enhance the ferroelectric properties as well as to improve the fatigue resistance [2-3]. For instance, Kan et al [4] showed that the dissipation factor of lanthanum doped-bismuth titanate was successfully reduced by about 8-10 times lower than of pure bismuth titanate. Besides that, the polarization properties of bismuth titanate material were greatly improved with the addition of neodymium [5]. In this work, however, the trivalent cation of neodymium (Nd^{3+}) is used as a substitution element in bismuth titanate compound in order to study the potential use as dielectric material for wireless application. In order to be applied in such field dielectric ceramic materials must have a high dielectric constant and low dielectric loss [1]. In our present study, a combustion route synthesis without fueling agent is used as it is simple, quick and easy which is of high significance for future robust industrial production [6].

II. METHODOLOGY

Bismuth titanate (is referred as undoped sample) and neodymium doped-bismuth titanate (is referred as doped sample) compounds with different compositions of neodymium (x=0.2Nd, 0.6Nd and 1.0Nd) were synthesized via fuel-free combustion route. Bismuth nitrate penthahydrate and neodymium nitrate hexahydrate were initially dissolved in 2-Methaoxyethanol (2-ME) at 40°C and stirred for about 30 min. Separately, titanium (IV) isopropoxide was dissolved in a homogeneous solution of 2-ME and acetylacetone (AA) at room temperature and stirred for 30 min. Then, both were mixed each other in a beaker. The solution temperature was then fixed at 40°C (±5) and stirred for another 2hrs. During this period, the solution slowly evaporated and formed in viscous solution. The temperature was then increased up to 130°C for the gel formation. The ignition and combustion temperature were recorded at 130°C and 200°C, respectively. Fluffy powder was formed after complete combustion within few minutes. The process was consistently repeated for others composition. The resultant powders were calcined at 800°C for 3 hrs. The calcined powders were then pressed into pellet, and directly sintered in air at 1100°C for 3 hrs. Phase formation, microstructure and dielectric properties of samples were then characterized by XRD (Bruker D8 Advanced), SEM (Zeiss Supra 55VP PGT/HKL Field Emission Scanning Electron Microscope) and Precision LCR meter (Model 4284A, Agilent), respectively. The process flow for the preparation of undoped and doped samples is shown in Fig. 1.

III. DIELECTRIC MEASUREMENT

The dielectric constant can be determined using the following equation [7].

$$\varepsilon = \varepsilon_o \varepsilon_r = \frac{t}{A} C_p$$

Where,

ε Dielectric constant (permittivity) [F/m]
ε_o Space permittivity = 8.854×10^{-12} [F/m]
ε_r Relative dielectric constant (Relative permittivity) of sample

C_p Equivalent parallel capacitance value [F]
t Thickness of sample [m]
A Area of electrode [m^2]

Thus, the relative dielectric constant (generally called the dielectric constant) of the test material, ε_r, can be obtained by measuring the capacitance value, calculated from the following equation.

$$\epsilon_r = \frac{t \times C_p}{A \times \epsilon_o} = \frac{t \times C_p}{\pi \times \left(\frac{d}{2}\right)^2 \times \epsilon_o}$$

Where,
d diameter of electrode [m]

The dissipation factor loss (= tan δ; loss tangen) of sample can be obtained directly by measuring the dissipation factor.

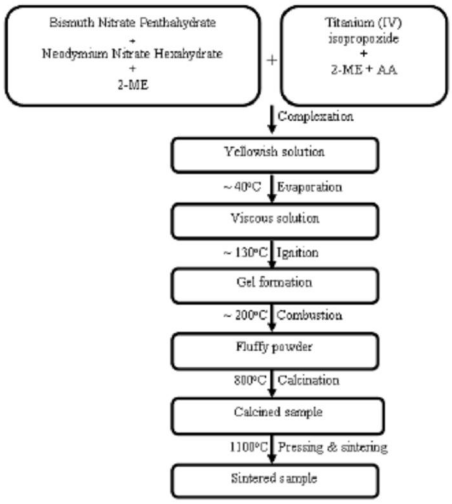


Fig. 1. Process diagram for the preparation of undoped and doped sample via fuel-free combustion route.

IV. RESULTS AND DISCUSSIONS

The XRD spectrum of undoped and doped samples at 800°C for 3 hrs are shown in Fig. 2. It was found that small amount of $Bi_{12}TiO_{20}$ intermediate phase was formed in the undoped sample, while such intermediate phase was completely vanished in the doped samples. It indicated that the addition of neodymium into bismuth titanate compound has reduced the formation of this intermediate phase.

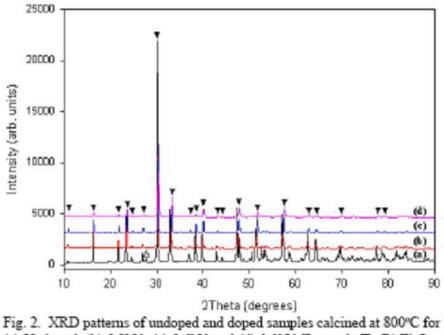


Fig. 2. XRD patterns of undoped and doped samples calcined at 800°C for 3h: (a) Undoped, (b) 0.2Nd, (c) 0.6Nd and (d) 1.0Nd [Legend: ▼: $Bi_4Ti_3O_{12}$ and Δ: $Bi_{12}TiO_{20}$].

Fig. 3 shows the SEM images of undoped and doped samples (specifically denoted as 1.0Nd) calcined at 800°C for 3 hrs. It can clearly be seen that undoped sample exhibits a plate-like structure and the grain size being over 1μm in length as shown in Fig. 3a. On the other hand, a remarkable decrease in the grain size with less than 1μm was observed in doped sample (Fig. 3b). From the size comparison between undoped and doped samples, it is reasonable to say that the substituted neodymium act as a grain-growth inhibitor in bismuth titanate-type compounds.

Both samples were then sintered at 1100°C for 3hrs and their images are shown in Fig. 4. Large rectangular grains of over 10μm in length were frequently observed in undoped sample (Fig. 4a). Upon increasing the neodymium content, those grains had reduced their size to be less than 10μm shown in Fig. 4b.

Fig. 5 shows the frequency dependences of dielectric constant (ε_r) and dissipation factor (tan δ) of the undoped and doped samples (specifically denoted as 1.0Nd) at room temperature. The electric field was set at 1V. Undoped sample had a strong dependence on test frequency and displayed a lower dielectric constant, ε_r, with the range of 152 - 161. With neodymium addition the dielectric constant of bismuth titanate, ε_r, increased gradually to be in the range of 173 – 183.

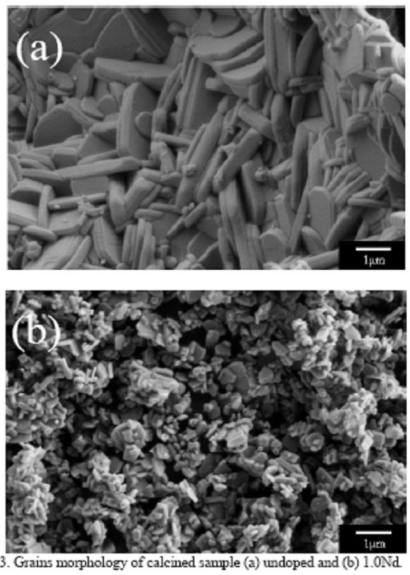


Fig.3. Grains morphology of calcined sample (a) undoped and (b) 1.0Nd.

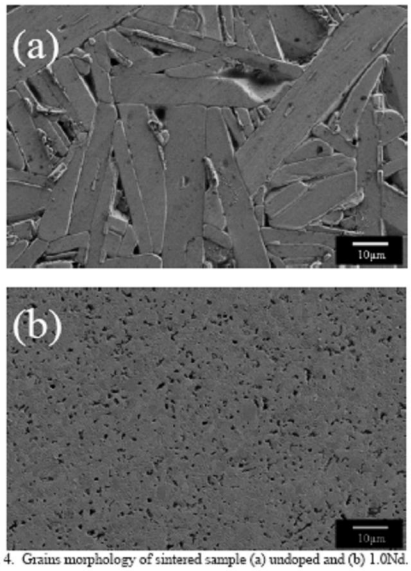
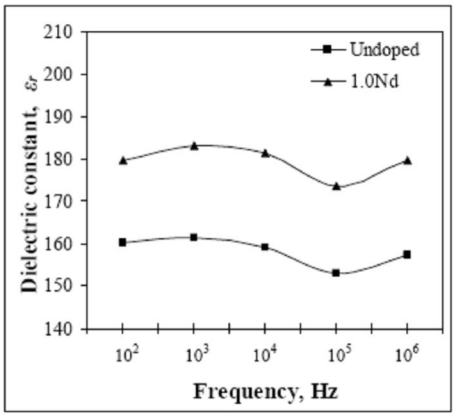
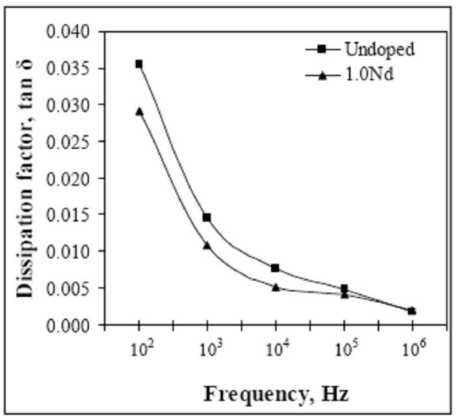


Fig. 4. Grains morphology of sintered sample (a) undoped and (b) 1.0Nd.



(a)



(b)

Fig. 5. Dielectric properties in dependence of frequency for undoped and 1.0Nd measured with an electric field of 1V: (a) dielectric constant, ε_r and (b) dissipation factor, tan δ.

Goh et al. [8] stated that the electrical properties of ferroelectric materials are highly dependent on grain size and microstructure of the samples. They claimed that large grain size (>1μm) had higher possibility producing high dielectric constant and it is attributed by internal stress and number of domain per grain.

However, their finding is contrast with the result obtained in present study. The fact that the dielectric constant increases with raising neodymium content proves that grain size and microstructure are not the only reason for changes in the dielectric constant. Simóes et al. [9] reported that the dielectric properties with frequency dependence are highly attributed by ionic defect e.g. trapped oxygen and combination of new defect.

Meanwhile, the dissipation factor of 1.0Nd doped sample is relatively low at about 1 to 1.5 times lower than of undoped sample. Such result is in agreement as per claimed in literature part indicating that the range of dissipation factor had reduced with the addition of neodymium into bismuth titanate compound.

IV. CONCLUSSION

One of the important aspects of the present work is to provide a processing method which is simple, quick and easy which is of high significance for future robust industrial production. To date, all the combustion synthesis uses fuel agent such as citric acid, urea or glyzine to assist the combustion process to form the powder in a finer size. In here we have proved that bismuth titanate and neodymium doped-bismuth titanate powder can be successfully synthesized without any addition of fuel. Other than that the experimental setup are simple and requires less than several hours as compared hydrothermal and wet chemical method.

ACKNOWLEDGMENT

The authors appreciate the technical support provided by the School of Materials and Mineral Resources Engineering, USM. This research was supported by the E-science Fund 305/Pbahan/6013357 and the USM Fellowship

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