

# Microstructure Characteristics and Electrical Properties of Sintered (Bi,La)<sub>4</sub>Ti<sub>3</sub>O<sub>12</sub> Ferroelectric Ceramics

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## Abstract

Imm-thick BLT ceramics were sintered in accordance with a bulk ceramic fabrication process. All XRD peaks detected in the sintered ceramics were indexed as the Bi-layered perovskite structure without secondary phases. Density was increased with increasing the sintering temperature up to 1050 C and the maximum value was about 98% of the theoretical density. The remanent polarization (2Pr) value of BLT ceramic sintered at 1050 C was approximately 6.5  $\mu$ C/cm<sup>2</sup> at the applied voltage of 4.5kV. From these results, a BLT ceramic target for plused laser deposition (PLD) system was successfully fabricated.

# Keywords : (Bi,La)<sub>4</sub>Ti<sub>3</sub>O<sub>12</sub>, Ferroelectric ceramics, Sintering, Microstructure, Electrical properties

# 1. Introduction

We Several ferroelectric materials including (Pb,Zr)TiO<sub>3</sub> (PZT) and (Bi,La)<sub>4</sub>Ti<sub>3</sub>O<sub>12</sub> (BLT) have been intensively studied for commercial applications such as nonvolatile ferroelectric random access memory (FeRAM) [1,2]. PZT has the advantages of low crystallization temperature ( $<650^{\circ}$ C) and large polarization value (2Pr>40  $\mu$ C/cm<sup>2</sup>) [1]. Nevertheless, environmental safety issues related with the Pb element may ultimately prevent it from being used in many applications. Ferroelectrics with bismuth-layered structure containing BLT have been thought to be promising materials for replacing the Pb-based ceramics [2,3]. The ferroelectric BLT material has many advantages in comparison with the most famous PZT-based ferroelectric materials: being fatigue free, having stable imprint characteristics and so on [2-3]. In addition, BLT is the Pb-free ecological material.

The BLT material is well known to have a strong anisotropy of the ferroelectric properties [4-6]. The remanent polarization of it is about 4  $\mu$ C/cm<sup>2</sup> along c-axis but the value was much large about 50  $\mu$ C/cm<sup>2</sup> along a-axis. Therefore, the orientation control of the BLT thin film has been one of the major issues for commercial applications.

We reported here on the fabrication processes of the BLT target for the PVD methods and also discussed microstructure and electrical properties of the sintered BLT ferroelectric ceramics.

#### 2. Experimental and Results

Fig. 1 shows the XRD patterns of the BLT ceramics

sintered at various temperatures. All of the samples were calcined at the same process condition of  $750^{\circ}C/4hr/Air$ . The sintering temperature was varied at the range of 900~1100°C. All of the detected XRD peaks were indexed as those of the Bi-layered perovskite structure without secondary phases [5]. The microstructure of the sintered BLT ceramics was inspected with the SEM equipment.

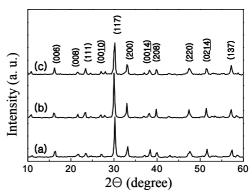


Fig. 1. XRD patterns of the BLT ceramics sintered at various temperatures: (a) 900 ℃, (b) 1000 ℃, (c) 1100 ℃.

Fig. 2 is the SEM images observed at the fractured surface. The samples sintered below  $1000^{\circ}$ C was not well densified. In contrast, the sample sintered at  $1100^{\circ}$ C was well densified.

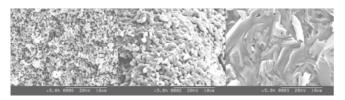


Fig. 2. SEM images of the BLT ceramics sintered at various temperatures.

Fig. 3 is the measured density of the BLT ceramics sintered at the various temperatures. The increasing of the density below  $1050^{\circ}$ C may be related with the densification of powders. And the decreasing of the density over  $1050^{\circ}$ C may be related with the volatility of the bismuth element. The maximum density was 7.5 g/cm<sup>3</sup>, which was 98% of the theoretical density, at the sample sintered at  $1050^{\circ}$ C.

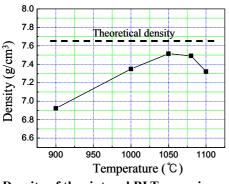


Fig. 3. Density of the sintered BLT ceramics.

Ferroeectric property was measured with the Precision-LC ferroelectric tester. Fig. 4 is the ferroelectric hysteresis loops of the sample sintered at 1050°C. The remanent polarization (2Pr) measured at the applied voltage of 4.5kV was approximately 6.5  $\mu$ C/cm<sup>2</sup> and the coecive field (2Ec) was about 52 kV/cm.

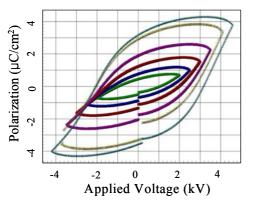


Fig. 4. Polarization-voltage hysteresis loops of the BLT ceramics sintered at 1050℃.

Piezoelectric parameters were also evaluated by using HP4294A impedence analyzer. Fig. 5 shows the impedance-frequency curve obtained at the thickness vibration mode. The relative dielectric constant was measured about 150. The calculated electromechanical couping factor ( $k_t$ ) of it was about 5% and the mechanical quality factor (Qm) was about 2200 [7].

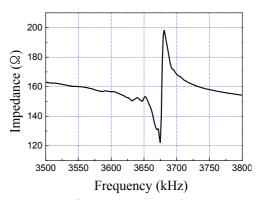


Fig. 5. Impedance-frequency curve of the BLT ceramics sintered at 1050℃.

## 3. Summary

BLT ceramics were sintered in accordance with bulk ceramic fabrication processes. All XRD peaks were indexed as the Bi-layered perovskite structure without secondary phases. Density was increased with increasing the sintering temperature up to 1050 °C and the maximum value was about 98% of the theoretical density. The remanent polarization (2Pr) value of BLT ceramic sintered at 1050 °C was approximately  $6.5\mu$ C/cm<sup>2</sup> at the applied voltage of 4.5kV. The calculated electromechanical quality factor (kt) of it was about 5% and the mechanical quality factor (Qm) was about 2200. From these results, a BLT ceramic target for PLD system was successfully fabricated. In the future, we will study on the thin film deposited with PLD system by using the successfully developed BLT ceramic target.

### 4. References

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