



## Original Research Paper

Preparation and characterization of the bismuth sodium titanate ( $\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3$ ) ceramic doped with ZnOChuen-Shii Chou<sup>a,\*</sup>, Chun-Yu Wu<sup>a</sup>, Ru-Yuan Yang<sup>b</sup>, Cheng-Yang Ho<sup>a</sup><sup>a</sup> Powder Technology R&D Laboratory, Department of Mechanical Engineering, National Pingtung University of Science and Technology, Pingtung 912, Taiwan<sup>b</sup> Department of Materials Engineering, National Pingtung University of Science and Technology, Pingtung 912, Taiwan

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## ABSTRACT

This study investigates effects of the zinc oxide (ZnO) addition and the sintering temperature on the microstructure and the electrical properties (such as dielectric constant and loss tangent) of the lead-free piezoelectric ceramic of bismuth sodium titanate ( $\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3$ ), NBT, which was prepared using the mixed oxide method. Three kinds of starting powders (such as  $\text{Bi}_2\text{O}_3$ ,  $\text{Na}_2\text{CO}_3$  and  $\text{TiO}_2$ ) were mixed and calcined. This calcined NBT powder and a certain weight percentage of ZnO were mixed and compressed into a green compact of NBT–ZnO. Then, this green compact of NBT–ZnO was sintered to be a disk doped with ZnO, and its characteristics were measured. In this study, the calcining temperature was 800 °C, the sintering temperatures ranged from 1000 to 1150 °C, and the weight percentages of ZnO doping included 0.0, 0.5, 1.0, and 2.0 wt%. At a fixed wt% ZnO, the grain size increases with increase in the sintering temperature. The largest relative density of the NBT disk obtained in this study is 98.3% at the calcining temperature of 800 °C, the sintering temperature of 1050 °C, and 0.5 wt% ZnO addition. Its corresponding dielectric constant and loss tangent are 216.55 and 0.133, respectively.

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## 1. Introduction

The piezoelectric material (such as lead zirconate titanate (PZT) system piezoelectric ceramic) has been attracted substantial interest because of its effective use in electronic components, such as the surface-wave filter, the piezoelectric transformer, and so on. However, the lead-free piezoelectric ceramic (such as  $\text{BaTiO}_3$ , or  $\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3$ ) is expected to replace PZT due to its low environmental impact. Comparing with  $\text{BaTiO}_3$ , the NBT ( $\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3$ ) has received more attention because the  $\text{BaTiO}_3$  has a relatively high-sintering temperature of 1350 °C approximately.

Recently, several methods (such as the hydrothermal synthesis [1], the stearic acid gel method [2], the citrate method [3], and the mixed oxide method [4]) have been adopted to fabricate the lead-free piezoelectric ceramic of NBT. Although the NBT, which is a perovskite-type ferroelectric, has a relatively large remnant polarization ( $P_r = 34 \mu\text{C}/\text{cm}^2$ ) at room temperature and a relatively high Curie temperature (320 °C) [3,5], its relatively high coercive field (73 kV/cm) makes the problem of poling NBT.

In order to solve the aforementioned problem, the additive (such as  $\text{Nd}_2\text{O}_3$  [6],  $\text{Eu}_2\text{O}_3$  [7],  $\text{MnCO}_3$  [8],  $\text{La}_2\text{O}_3$  [9],  $\text{Sm}_2\text{O}_3$  [9],  $\text{Nb}_2\text{O}_5$  [9],  $\text{Fe}_2\text{O}_3$  [10], or  $\text{Dy}_2\text{O}_3$  [11]) was added in the NBT to

modify and enhance the relative properties (such as grain size, relative density, etc.) of the NBT. Chou et al. [12] observed that the NBT disk doped with MgO not only improved the relative density of the disk, but also lowered the sintering temperature. Moreover, Chou et al. [13] used mixed oxide method and the Taguchi method [14] to optimize the conditions of preparing the disk of NBT without adding any additive.

However, adding ZnO in the NBT has seldom been studied, although ZnO additive was successfully added in the other kinds of ceramic (such as  $\text{La}_{0.67}\text{Ca}_{0.33}\text{MnO}_3$  [15],  $\text{Zr}_{0.8}\text{Sn}_{0.2}\text{TiO}_4$  [16],  $\text{BiNbO}_4$  [17],  $\text{BaZr}_x\text{Ti}_{1-x}\text{O}_3$  [18],  $\text{BaTiO}_3$  [19],  $\text{SnO}_2$  [20],  $\text{Zr}_{0.8}\text{Sn}_{0.2}\text{TiO}_4$  [21], and  $\text{LaAlO}_3$  [22]). It has been generally agreed with that ZnO can promote the densification and control the grain growth during sintering process [18]. Aside from these references, Lee et al. recently synthesized ZnO-doped NBT by the conventional solid state reaction method and presented several useful information of ZnO-doped NBT (such as piezoelectric coefficient and electromechanical coupling factor) [23]. They indicated that the ZnO assisted in the densification of the NBT ceramics by promoting liquid phase sintering (LPS), but less attention has been paid to how the LPS occurred and affected the sintering of NBT. Accordingly, the research on the ZnO-doped NBT is worthy of continuing study.

Therefore, the objective of this study is to investigate the mechanism of enhancing the relative density of a NBT–ZnO disk. Aside from this, the effects of the ZnO addition and the sintering temperature on

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