Physics Letters A 373 (2009) 1583–1587

Contents lists available at ScienceDirect

Physics Letters A

www.elsevier.com/locate/pla

Influences of compressive stress and aging on dielectric properties of sodium bismuth titanate ceramics

T. Sareein, M. Unruan, A. Ngamjarurojana, S. Jiansirisomboon, A. Watcharapasorn, R. Yimnirun *

Department of Physics, Faculty of Science, Chiang Mai University, Chiang Mai, 50200, Thailand

ARTICLE INFO

Article history:
Received 15 December 2008
Received in revised form 17 February 2009
Accepted 23 February 2009
Available online 5 March 2009
Communicated by A.R. Bishop

PACS:
77.80.-e
77.80.Fm
77. 84.-s

Keywords:
Dielectric properties
NBT
Compressive stress
Aging

ABSTRACT

The influences of compressive stress and aging on dielectric properties of undoped and Fe-doped sodium bismuth titanate (NBT) ceramics were investigated. The dielectric properties were decreased significantly with the compressive stress applied parallel to the electric field direction, while the changes were reversed with the stress applied perpendicularly. In addition, lower changes of the dielectric properties with stress were observed in Fe-NBT ceramics, likely caused by an enhanced relaxor characteristic with the acceptor doping, which also reduced the aging rate in the ceramics. Finally, the aging behavior of the NBT and Fe-NBT ceramics followed the slightly stretched exponential law, and the aging rate in both ceramics was found to decrease with frequency, a result of the pinning of the polarization components. These observations clearly confirmed the role of the acceptor dopant in enhancing the relaxor ferroelectric characteristics in the NBT-based ceramics.

1. Introduction

Sodium bismuth titanate, $Na_{0.5}Bi_{0.5}TiO_3$ (NBT), and NBT-based ceramics are currently considered as a potential lead-free ferroelectric material to replace the widely used lead-based perovskite materials because of lead-free control atmosphere and lack of lead pollution [1–10]. While several previous investigations have suggested that the NBT exhibits strong ferroelectric properties with a large remnant polarization, $P_r = 38\ \mu C/cm^2$, and a phase transition point from ferroelectric to antiferroelectric should occur above 200 °C, many experiments do not confirm this hypothesis [1,2,4,10]. At present researchers accept that rather coexistence of rhombohedral and tetragonal phases takes place above 200 °C, not antiferroelectric [11]. In addition, this material system has been investigated in terms of variation in their dielectric and piezoelectric properties due to processing conditions, various dopants, and the formation of solid solution with other compounds [2,4–7,9,10].

Piezoelectric and ferroelectric ceramics are widely used in devices such as actuators and transducers. However, when they are used in devices, these ceramics are often subjected to self-induced or under environmental stresses. A prior knowledge of the effects of stresses on the material properties is crucial for proper design of a device and for suitable selection of materials for a specific application. Therefore, it is very important to obtain experimental data, as well as to better understand how these materials behave under stress [12–15]. Recently, the compressive stress dependence of dielectric properties has been studied in lead-based ferroelectric materials [16–23]. Interestingly, there have been many previous reports on the electrical properties of NBT-based ceramics [4–9], but there has been no systematic study on the influence of an applied stress on the dielectric properties of the NBT ceramics. Earlier investigations have already revealed significance of stress-induced ferroelectric phase transition, and domain structure and birefringence in NBT single crystals and NBT-based ceramics [24–31]. In addition to practical implication of stress influence, aging, usually regarded as an unwanted effect [32–35], has also been a subject of interest in lead-free materials, as it has been reported that aging of piezoelectric properties of lead-free materials is dramatic, hence limiting their potential uses in many applications [36–42]. Though aging behavior has been investigated extensively in lead-based materials [33,43–47], the aging behavior of the dielectric properties of NBT-based ceramics has yet to be reported.

Therefore, this study is aimed to examine the dielectric properties of the $Na_{0.5}Bi_{0.5}TiO_3$ ceramics as a function of compressive stress applied parallel and perpendicular to an electric field direction. Aging behavior of the dielectric properties of the ceramics is also investigated. This study is also extended to Fe-doped NBT

* Corresponding author at: School of Physics, Institute of Science, Suranaree University of Technology, Nakhon Ratchasima 30000, Thailand.
E-mail address: rattikornyimnirun@yahoo.com (R. Yimnirun).

0375-9601/$ – see front matter
doi:10.1016/j.physleta.2009.02.065

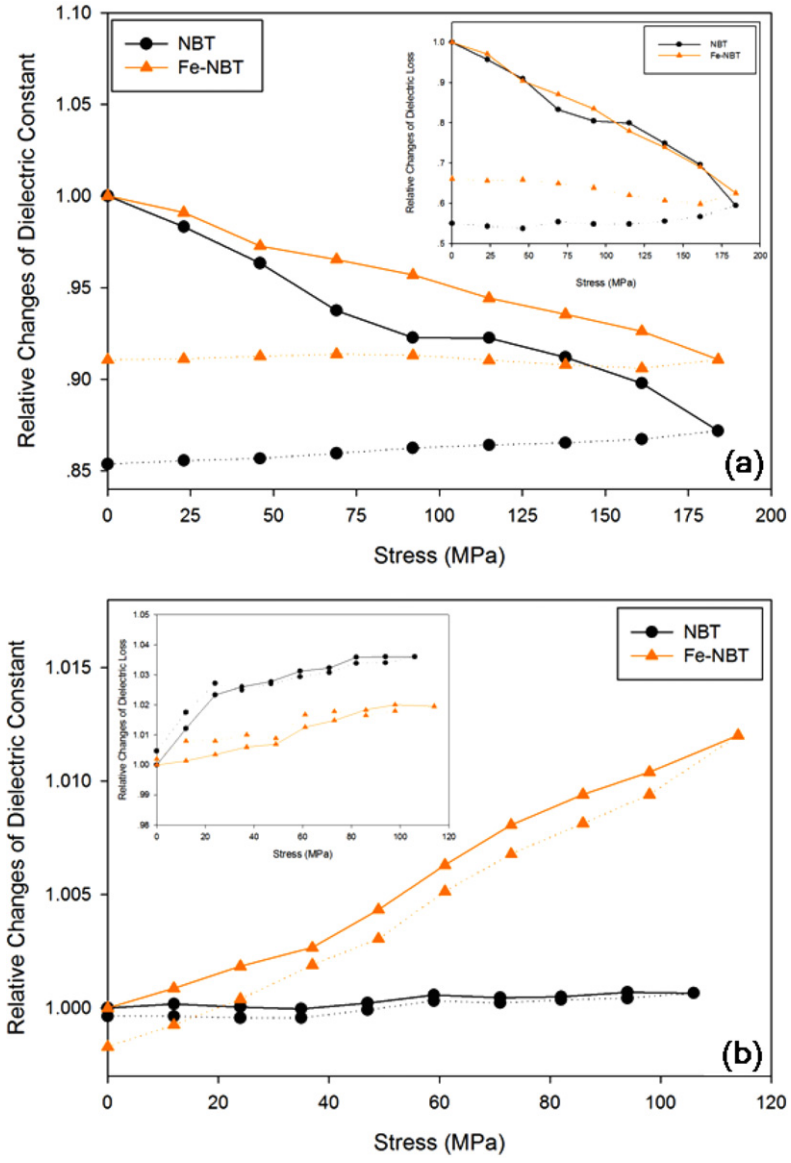

Fig. 1. Relative changes of dielectric constant (ε_r) and relative changes of dielectric loss tangent (tan δ) (inset) for NBT and Fe-NBT ceramics with compressive stress applied parallel (a) and perpendicular (b) to an electric field direction (measured at 25 °C and 10 kHz). Solid lines indicate changes during loading, and dotted lines indicate changes during unloading. Opposite trends are observed in the two stress directions.

ceramics since it is well established that lower-valence doping entails aging and previous investigations already showed significantly different responses to stress in undoped and doped (both lower-valence and higher-valence) lead-based materials [18,19,22,23]. Therefore, it is of very interest to also conduct the same experiments in the Fe-doped NBT ceramics.

2. Experimental

In this study, $Na_{0.5}Bi_{0.5}TiO_3$ (NBT hereafter) and Fe-doped $Na_{0.5}Bi_{0.5}TiO_3$ (Fe-NBT hereafter) powders were prepared from binary oxides and carbonate, i.e. Bi_2O_3 (>98%, Fluka), Na_2CO_3 (99.5%, Carlo Erba), TiO_2 (>99%, Riedel-de Haën) and Fe_2O_3 (99.9%, Fluka) with the nominal composition $Na_{0.5}Bi_{0.5}Ti_{1-x}Fe_xO_{3-0.5x}$, where $x = 0.0$ and 0.015. The powder mixtures were ball milled in ethanol using zirconia milling media for 24 h, calcined at 800 °C for 2 h. The calcined powders were re-ground using agate mortar and pestle, sieved, pressed into pellets and sintered at temperature of 1050 °C for 2 h. Detailed preparation process is given elsewhere [48]. The sintered specimens were then cut as rectangular bars (typical dimensions $6 \times 2 \times 2$ mm^3) and lapped to obtain parallel faces. After coating with silver paint as electrode, the effects of the compressive stress on the dielectric properties of the aged ceramics (the specimens were left at 25 °C for 4 weeks to achieve a fully aged state) were investigated with the compressometer. The compressive stress was applied both parallel and perpendicular to an electric field direction [49,50]. The low-field dielectric properties were measured by LCR-meter (Instrek LCR-821) with applied voltage of 1 V. The room temperature (25 °C) dielectric constant and dielectric loss tangent were determined at frequency 10 kHz. For aging study, the samples were heated to 550 °C and kept for 30 min to remove the aging effect and ensure them to start from unaged states. Then, they were immediately cooled to room temperature. The aging effect was characterized by measuring the changes in the dielectric properties with aging time.

3. Results and discussion

By X-ray diffraction method, both NBT and Fe-NBT ceramics were single phase with a rhombohedral structure. SEM micro-