

Processing and properties of CeO₂-doped ferroelectric (Na_{0.5}Bi_{0.5})_{0.94}Ba_{0.06}TiO₃

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Abstract

The (Na_{0.5}Bi_{0.5})_{0.94}Ba_{0.06}TiO₃–0.6 wt.%CeO₂ (abbreviated as NBT-BT6–0.6%CeO₂) ceramic was synthesized by a citrate method. The effects of poling condition on piezoelectric properties of NBT-BT6–0.6%CeO₂ ceramic and the influences of sintering temperature on its ferroelectric and piezoelectric properties were investigated. It was found that the piezoelectric properties of NBT-BT6–0.6%CeO₂ ceramic highly rely upon poling field and poling temperature, while no remarkable effect of poling time on the piezoelectric properties was found in the range of 5–25 min. Moderate increase of sintering temperature improves the piezoelectric and ferroelectric properties of NBT-BT6–0.6%CeO₂ ceramic. With respect to the piezoelectric and ferroelectric properties, a sintering temperature range of 1130–1170 °C is ascertained for NBT-BT6–0.6%CeO₂ ceramic. © 2005 Elsevier B.V. All rights reserved.

Keywords: (Na_{0.5}Bi_{0.5})_{0.94}Ba_{0.06}TiO₃–0.6 wt.%CeO₂ ceramic; Poling condition; Sintering temperature; Piezoelectricity; Ferroelectricity

1. Introduction

In recent years, lead-free piezoelectric ceramics have attracted considerable attention as an important piezoelectric material because of their outstanding advantages in high mechanical strength, lack of air pollution and free control of toxic atmosphere during sintering. Sodium bismuth titanate, (Na_{0.5}Bi_{0.5})TiO₃ (abbreviated as NBT), is a kind of perovskite-type ferroelectric with complex A-site cations, showing a relatively large remanent polarization ($P_r = 38 \mu\text{C}/\text{cm}^2$) at room temperature and a relatively high Curie temperature ($T = 320^\circ\text{C}$) [1]. For its strong ferroelectricity at room temperature, NBT has been considered to be a promising candidate material for lead-free piezoelectric ceramics. However, the wide applications of NBT have been limited for its high coercive field and the high conductivity. Coercive field of NBT is very close to its breakdown voltage, which prevents the complete domain reversal under high electric field of the poling process. High

conductivity of NBT is also undesirable for the dielectric and piezoelectric application. To solve these problems, various NBT-based solid solutions have been developed [2–9]. Among them, (Na_{0.5}Bi_{0.5})_{1-x}Ba_xTiO₃ is more interesting owing to the existence of a rhombohedral (F_α)–tetragonal (F_β) morphotropic phase boundary (MPB). The further enhancement on the piezoelectric and ferroelectric properties of

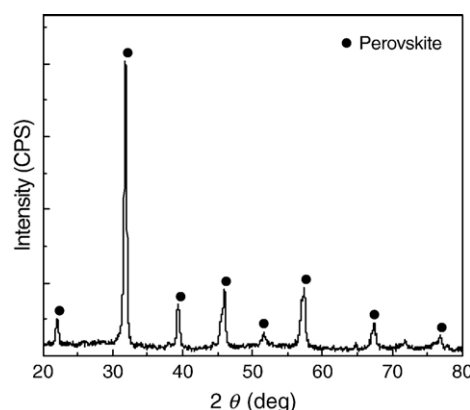


Fig. 1. XRD pattern of NBT-BT6–0.6%CeO₂ ceramic sintered at 1150 °C.

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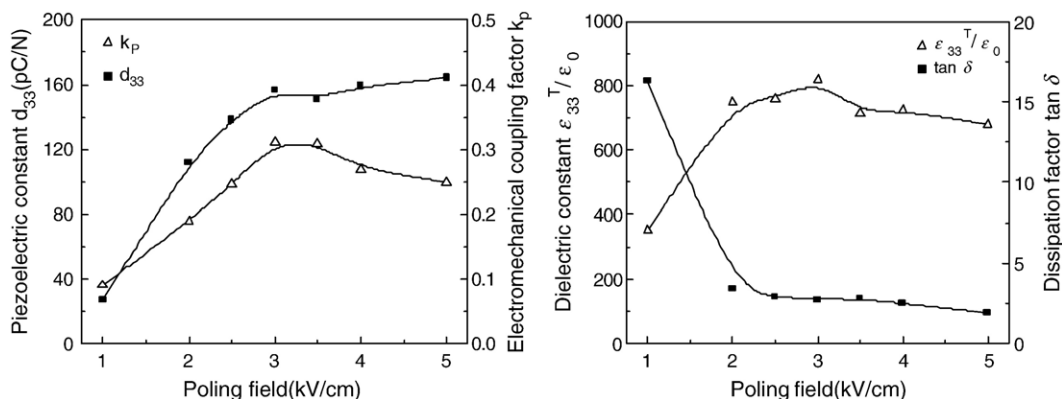


Fig. 2. Dependence of piezoelectric properties on poling field for NBT-BT6–0.6%CeO₂ ceramic sintered at 1150 °C. The specimens were poled at 60 °C for 15 min.

(Na_{0.5}Bi_{0.5})_{0.94}Ba_{0.06}TiO₃ (abbreviated as NBT-BT6) ceramic is reported by X. X. Wang [10] and H.D. Li [11].

Recently, chemical method is given attention and adopted to synthesize NBT based ceramics. The poling and sintering are requisite processes to yield desired electrical properties. Therefore, it is necessary to examine the electrical properties of NBT-BT6–0.6%CeO₂ ceramic synthesized by citrate method with respect to its poling condition and sintering temperature. Several previous researches involve the effect of poling condition or sintering temperature on the piezoelectric properties of NBT based ceramics [3,5,12]. However, the investigation into this subject is still limited. In our experiment the resulting (Na_{0.5}Bi_{0.5})_{0.94}Ba_{0.06}TiO₃–*x*wt.%CeO₂ (*x*=0, 0.2, 0.4, 0.6, 0.8, 1.0) ceramics by a citrate method exhibit superior piezoelectric and ferroelectric properties at *x*=0.6 which will be involved in other paper. So in the present work, the NBT-BT6–0.6%CeO₂ ceramic was synthesized by a citrate method. The effects of poling condition on piezoelectric properties of NBT-BT6–0.6%CeO₂ ceramic and the influences of sintering temperature on its ferroelectric and piezoelectric properties were investigated.

2. Experimental procedures

The citrate method was used to prepare NBT-BT6–0.6%CeO₂ powders. Reagent grade NaNO₃, Bi(NO₃)₃·5H₂O, Ba

(NO₃)₂, Ce(NO₃)₃·6H₂O, tetrabutyl titanate and citric acid were used as starting materials. Citric acid was first dissolved into deionized water in a beaker. After adjusting the pH value of the solution to 7–9 by dripping an appropriate amount of ammonia solution, tetrabutyl titanate was added under a stirring at 60 °C. An hour later, a yellowish two-layer liquid was obtained, comprising a transparent aqueous solution at the lower layer and an oil-like liquid at the upper layer. The aqueous solution was separated from the mixing liquid. Then various nitrates were added into the solution one after another, followed by stirring at 80 °C for 1 h to generate a precursor solution with a pH value of about 6. The precursor solution was dehydrated in an oven at 100 °C to form a sol. Subsequent heating at a higher temperature of 160 °C yielded a gel. The gel was pulverized and then calcined at 550–700 °C for 1 h in air. The calcined powders were granulated with PVA as a binder. The granulated powders were pressed into discs in diameter of 19 mm and thickness of 1 mm. The compacted discs were sintered at 1100–1180 °C for 2 h in air.

The crystal structure of NBT-BT6–0.6%CeO₂ ceramic was examined by a Rigaku D/MAX-RB X-ray diffractometer using CuK_α radiation. The microstructure of the specimens was observed by the Sirion FEG (FEI, USA). Silver paste was fired on both faces of the discs at 650 °C as electrodes. The specimens for measurement of piezoelectric properties were poled in silicon oil under different poling conditions. After poling, the piezoelectric

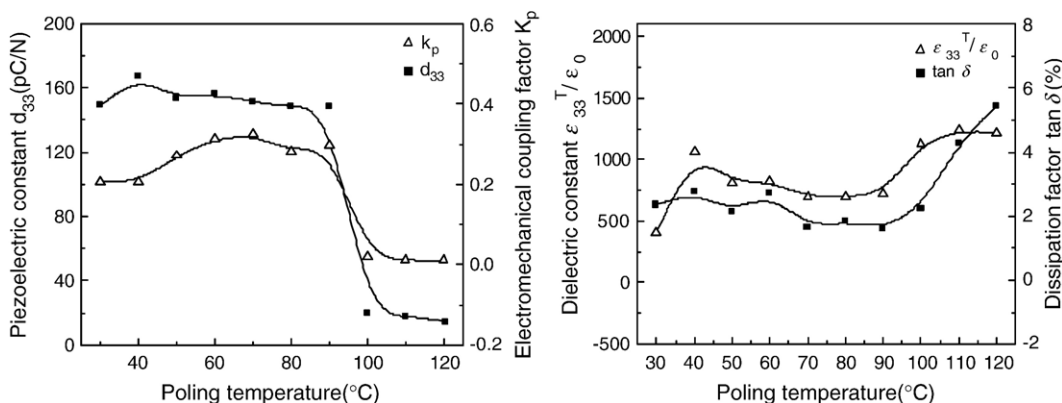


Fig. 3. Dependence of piezoelectric properties on poling temperature for NBT-BT6–0.6%CeO₂ ceramic sintered at 1150 °C. The specimens were poled under 3.0 kV/mm for 15 min.

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