Effects of quenching on bending strength and piezoelectric properties of (Bi$_{0.5}$Na$_{0.5}$)TiO$_3$ ceramics

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ABSTRACT

Lead-free piezoelectric bismuth sodium titanate (Bi$_{0.5}$Na$_{0.5}$)TiO$_3$ (BNT) ceramics were prepared by quenching and the correlation among a depolarization temperature $T_d$, the mechanical strength, and their piezoelectric properties were investigated. We controlled the quenching rate in the temperature range from 1100°C to 800°C. We examined the 5 patterns of the quenching rate (QR): 0.05 (ordinary firing, OF), 3.85, 5.77, 6.67, and 15.0 (quenching). $T_d$ of the rapid quenched sample reached 229°C (QR = 15.0), but its mechanical strength decreased to a quarter of that of the OF ceramics. On the other hand, $T_d$ was 211°C at QR = 3.85 which was higher than that of OF-ceramic (QR = 0.05) at about 180°C. Moreover, three-point bending strength (227 MPa) of the quenched BNLT4 (QR = 3.85) was equivalent to that of the OF ceramics (235 MPa). Therefore, both $T_d$ and the mechanical strength were improved by controlling the quenching rate.

1. Introduction

Piezoelectric ceramics play a very important role in contemporary electronic devices. Many piezoelectric devices are composed of Pb(Zr,Ti)O$_3$ (PZT)-based piezoelectric solid-solution systems owing to their outstanding piezoelectric characteristics [1–3]. However, piezoelectric materials containing lead are beginning to be regulated by laws such as Restriction of Hazardous Substances (RoHS) in order to reduce environmental pollution [4]. The commercial market for piezoelectric devices is dominated by actuators and high-power piezoelectric applications. Thus, the development of lead-free piezoelectric materials is urgently required. As candidates for lead-free piezoelectric and ferroelectrics materials, BaTiO$_3$ [5–9], (Bi,Na)TiO$_3$ [10–21], (K,Na)NbO$_3$ [22–32], and their solid-solution systems have been reported.

Bismuth sodium titanate (Bi$_{0.5}$Na$_{0.5}$)TiO$_3$ (BNT) is a promising lead-free piezoelectric material with a perovskite structure of rhombohedral symmetry (R3c) at room temperature (RT) [18,33,34]. BNT exhibits strong ferroelectric properties and its remanent polarization $P_r$, of 38 μC/cm$^2$ is relatively large. The piezoelectric properties of BNT-based solid-solution systems have been extensively studied and BNT-based compositions with morphotropic phase boundaries (MPBs) exhibited relatively large piezoelectric strain constants $d_{33}$ [11–16,22,35,36]. Additionally, BNT-based ceramics show a stable mechanical quality factor $Q_m$ even under high-amplitude vibration, and are suitable for high-power piezoelectric applications such as ultrasonic devices [37]. Therefore, BNT-based ceramics are promising lead-free piezoelectric materials because they are compatible with current manufacturing processes and have a low material cost and relatively good piezoelectric properties. However, BNT exhibits a low depolarization temperature $T_d$ of ~180°C. That is, its range of operating temperatures is narrow.

In recent years, quenching during sintering has been proposed, and quenching above 800°C has been reported to be effective for increasing $T_d$ of pure BNT ceramics without deteriorating their ferroelectric and piezoelectric properties [38]. The quenching is effective above 800°C and does not affect at temperatures below 600°C. That is, since the structural phase transition of BNT occurs below 600°C, the quenching effect is independent of the structural phase transition. $T_d$ of BNT ceramics quenched from 1100°C was 223°C, which was about 50°C higher than that of ceramics prepared by the ordinary cooling process. Moreover, it was elucidated that the quenching procedure enhanced the lattice distortion of the rhombohedral phase (90-α) and provided a BNT ceramic with a higher $T_d$. Yoneda et al. also reported such enhancement of the rhombohedral distortion in Li-substituted BNT (BNLT) ceramics [39]. $T_d$ of BNLT with Li substitution was ~20°C higher than that of pure BNT [40]. Miura et al. achieved a ~50°C higher $T_d$ of...
BNLT by using a quenching process [41]. Then, \((1-x)\)BNT–\(x(Bi_{0.5}Li_{0.5})TiO_3\) (BNLT100x) with \(x = 0.04\) showed a higher \(T_d\) of 245°C after quenching treatment.

However, it is well known that quenched ceramics are generally broken or chipped easily even though their mechanical strength is important for their application to piezoelectric devices. For piezoelectric applications, it is essential to examine the mechanical strength because a large stress of 100 MPa or more is induced in a piezoelectric sample at a vibration velocity of 1 m/s on ultrasonic devices for example [42]. So far, many studies have been performed on the mechanical properties of PZT ceramics [43–47]. The bending strengths of various PZT-based ceramics range from 50 to 150 MPa [43–47]. On the other hand, according to some reports, the bending strengths of BNT-based ceramics are 150–200 MPa [7,48,49]. Moreover, it has been reported that the mechanical strength of BNT-based ceramics can be increased to over 200 MPa by controlling the grain size, additives such as Nb and processing such as hot pressing [50]. In these reports, the bending strength of BNT-based ceramics is higher than that of PZT-based ceramics. For this reason, BNT-based ceramics are suitable as a lead-free piezoelectric material for piezoelectric applications. However, the mechanical strength of the quenched BNT has not yet been clarified. Therefore, in order to maintain both high \(T_d\) and large mechanical strength, it is necessary to examine the mechanical strength of samples of quenched BNT. In this study, we controlled the temperature gradient from 1100°C to 800°C during quenching. Furthermore, we investigated the correlation between \(T_d\) and the mechanical strength, and the piezoelectric properties of the quenched samples.

2. Experimental methods

0.96(Bi_{0.5}Na_{0.5})TiO_3-0.04(Bi_{0.5}Li_{0.5})TiO_3 (BNLT4) ceramics were prepared by a conventional ceramic fabrication technique. Reagent-grade metal oxide or carbonate powders of Bi_2O_3, TiO_2, Na_2CO_3, and Li_2CO_3 were used as raw materials. The ceramic fabrication procedure is able to be found elsewhere [41]. Calcination was carried out at 850°C for 2 h, which was followed by sintering at 1140°C for 2 h in an alumina crucible. The sintering program is schematically shown in Figure 1. After sintering, the ceramics were cooled at a rate of 200°C/h in the OF process and are hereafter referred to as OF-BNLT4.

On the other hand, after sintering, some samples were removed from the electric furnace set at a high temperature of 1100°C, exposed to room air, and cooled rapidly to RT using a water bath, silicone oil bath or air fan.

Figure 2(a) shows temperature gradients from 1100°C to 800°C in each QR for OF and quenched ceramics. These gradients were controlled by intensity of air fan. This is because the temperature range from 1100°C to 800°C is effective in quenching [38]. In the range, the temperature gradient for quenching was controlled using the following formula:

\[
\text{Quenching rate (QR)} = \frac{1100 - 800}{t_q},
\]

where \(t_q\) is the time interval taken for the temperature to fall from 1100°C to 800°C. QR was set to five values: 0.05 (OF), 3.85, 5.77, 6.67, and 15.0°C/s as shown in Figure 2(b).

The densities of the ceramics were measured by the Archimedes method. The ceramics were processed for
various physical and electrical measurements. The crystal structure and lattice constant were observed by X-ray diffraction using Cu Kα radiation (XRD: Rigaku RINT-2000, 40 kV, 40 mA) on the surface of each bulk specimen. Gold electrodes were fabricated on each ceramic by sputtering for the measurement of dielectric and piezoelectric properties. The longitudinal vibration in the (33) mode was measured using a rectangular solid specimen of $2 \times 2 \times 5 \text{ mm}^3$. The samples for piezoelectric measurement were poled in a silicone oil bath by applying a dc electric field of 5 kV/mm for 5 min at RT. The piezoelectric properties were evaluated by the resonance – antiresonance method in accordance with IEEE standards using an impedance analyzer (HP4294A). The temperature dependences of the same setup of the impedance analyzer were evaluated from RT to about 350°C. Then, the depolarization temperature $T_d$ was determined from the temperature dependences of the dielectric constant $\varepsilon_r$ and loss tangent $\tan \delta$.

Moreover, the mechanical strength of 12.0 × 4.0 × 1.0 mm³ samples was measured by performing three-point bending test based on JIS. At least 10 samples were examined. A load was applied to the samples moving at a speed of 0.5 mm/min and subsequently, the applied load was measured at the time when the samples fractured. The fracture stress was estimated from the obtained values of the applied load. Furthermore, the fracture probability was calculated and the relationship between the probability and the stress was plotted corresponding to a Weibull plot. On the basis of the Weibull plot, the stress value probability of 50% was defined as the average fracture stress $\sigma_A$, and the slope obtained by linear approximation of the plot, that is, the Weibull modulus, was determined. Microstructure of these samples was observed using scanning electron microscope (SEM) (Hitachi, S-2400). For preparing SEM samples, the surface of the samples was polished with 1 μm diamond paste, and then were thermally etched at 1090 °C for 1 h. An average grain size was obtained using a line intercept method.

### 3. Results and discussion

#### 3.1. The influences of cooling procedures

For all the samples, the relative densities of the prepared ceramics were high 98% or more of the theoretical density. Bulk BNT samples of about 17 mm diameter and 15 mm height were quenched using three methods: water cooling, cooling in a silicone-oil bath, and air cooling. After the samples were quenched, these samples were sliced into disks of 0.5 mm. Subsequently, in the cases of water cooling and cooling in a silicone-oil bath, the samples were broken into many pieces, 3 mm and 7 mm on average, respectively, as shown in Figure 3. The sample cooled in a silicone-oil bath also had partly black because the sample with 1100°C was dunk into the silicone-oil bath and thereby partly charred. These samples could not be used. For example, it would be very difficult to process such samples into a specific shape or to polarize them for a specific vibrational mode. On the other hand, the air-cooled samples maintained their shape and were not broken into many pieces. That is, the air-cooled samples had the same shape as the samples subjected to ordinary firing (OF). However, even for the air-cooled samples, the mechanical strength average bending strength, $\sigma_A$ was decreased from 235 to 61 MPa, a quarter of that of the OF samples.

![Figure 3. Photographs of OF-BNT and BNT cooled by three methods.](image)

![Figure 4. XRD patterns of BNLT4 ceramics for each QR.](image)
3.2. $T_d$ and mechanical strength of BNLT4 controlled by QR

Figure 4 shows the XRD patterns of OF- and air quenched BNLT4 (q-BNLT4). These patterns were adjusted and normalized using the strongest peak, since the possibility that the reflection angle shifted slightly was considered because of the measurement of disk-like samples. The XRD patterns indicated a single-phase perovskite structure with rhombohedral structures. This means that no secondary phases were induced in these samples by quenching. Lattice parameters and lattice distortions were estimated for OF- and q-BNLT4 from these patterns. The average lattice constant was 3.90 Å and the similar values were obtained regardless of the values of QR. Figure 5 shows rhombohedral distortion, 90-$\alpha$, for OF- and q-BNLT4 as a function of QR. The values of 90-$\alpha$ increased with increasing QR from 0.368 to 0.409. As a result, the lattice distortion was enhanced by QR. This kind of change in lattice distortion was also observed in other BNT-based solid solution ceramics by quenching procedure [38,41,51]. The reason for this was described in a paragraph in Figure 8.

The piezoelectric properties (a) electromechanical coupling factor, $k_{33}$, (b) piezoelectric strain constant, $d_{33}$, and (c) dielectric constant, $\varepsilon_{33}/\varepsilon_0$ of OF- and q-BNLT4 as a function of QR are shown in Figure 6. $k_{33}$ remained constant regardless of QR and maintained of an average value of 0.46. Therefore, the piezoelectricity $k_{33}$ was not deteriorated by quenching. $d_{33}$ values of q-BNLT4 with the four values of QR were slightly lower than that of OF-BNLT4. Also, the QR dependence of $\varepsilon_{33}/\varepsilon_0$ was consistent with the behavior of $d_{33}$. Therefore, we consider that the slight decrease in $d_{33}$ is induced by the reduction in $\varepsilon_{33}/\varepsilon_0$. Also, we assumed that the reduction of $\varepsilon_{33}/\varepsilon_0$ is originated from the enhancement of rhombohedral distortion 90-$\alpha$ and the increase of the $T_d$ as described in next paragraph.

Figure 7(a,b) shows the temperature dependences of the dielectric constant $\varepsilon_s$ and loss tangent $\tan\delta$, respectively, for poled OF- and q-BNLT4 (QR = 3.85 and 15.0). The temperature dependences of $\varepsilon_s$ showed the maximum peak at ~300°C and an anomalous shoulder at a lower temperature. The maximum peak temperatures $T_m$ are almost constant for each QR. On the other hand, the temperatures of first shoulders ranged from 180°C to 229°C and corresponded to the maximum peak of $\tan\delta$ [36]. $T_d$ was obtained as the temperature of the maximum peak of $\tan\delta$. The $T_d$ values of OF- and q-BNLT4 (QR = 3.85 and 15.0) were 180°C, 211°C and 229°C, respectively. $T_d$ for QR = 3.85 was similar to that for QR = 15.0 and about 30°C higher than that of OF-BNLT4. Moreover, for these QRs, the lattice distortions were 0.368, 0.402, and 0.409, respectively, according to Figure 5. The $T_d$ values of all the BNLT4 samples as a function of QR are summarized in Figure 8. $T_d$ and the lattice distortion tended to increase with increasing QR. The strong correlation between $T_d$ and lattice distortion was mentioned in a previous report [38,41,51], although its mechanism is not clearly understood. A possible cause is not the internal stress but the
Figure 7. Temperature dependences of dielectric constant $\varepsilon_r$ and loss tangent tanδ for BNLT4 ceramics with QR of 0.05 (OF), 3.85 and 15.0 (quenching) °C/s.

Figure 8. Depolarization temperature $T_d$ as a function of QR.

Figure 9. (a) Weibull plots of three-point bending strength $\sigma$ and fracture probability $F$. (b) Average bending strength $\sigma_A$ as a function of QR.
BNTs with QR = 0.05 and 15.0 was approximately 2.5 μm. Therefore, the pore size is the same for both QR = 0.05 and 15.0. In Figure 10(b), the above microcracks were not observed.

4. Conclusion

The effect of quenching was investigated by controlling QR in the range from 1100°C to 800°C in order to increase the mechanical strength of BNLT4. \( T_d \) increased, even though QR was lower than that (15.0) studied so far. Furthermore, the mechanical strength \( \sigma_A \) at QR = 3.85 was equal to that of OF samples. Therefore, we found the condition to increase the \( T_d \) while maintaining the mechanical strength without deteriorating their piezoelectric properties. Quenched BNLT4 ceramics are suitable as a lead-free piezoelectric material for applications such as electronic devices.

Disclosure statement

No potential conflict of interest was reported by the authors.

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