Temperature dependence of unipolar fatigue behavior of lead-free alkali niobate ceramics

Alexander MARTIN,† Ken-ichi KAKIMOTO, Keiichi HATANO‡ and Yutaka DOSHIDA*  
Frontier Research Institute for Materials Science, Nagoya Institute of Technology, Gokiso-cho, Showa-ku, Nagoya 466-8555, Japan  
*Materials R&D Department, R&D Laboratory, TAIYO YUDEN CO., LTD., Takasaki, Gunma 370–3347, Japan

1. Introduction

Piezoelectric actuators demonstrate high efficiency, unlimited resolution and ultrahigh accelerations.1–3 This is because, for this reason, they can be found in many different applications such as fuel injectors, nanopositioners and pulse drive motors, which involve the coupling of mechanical strains with high electric AC fields. Under these conditions the material is susceptible to changes in their characteristics. It was shown that lead zirconate titanate (abbreviated as PZT), decreases in piezoelectric and dielectric properties. It was shown that lead zirconate titanate (abbreviated as PZT), decreases in piezoelectric and dielectric properties after a certain amount of electric cycles.4,5)

More information about the occurring processes that lead to these changes can be deduced from fatigue experiments. Given the widespread use of PZT in industrial applications, most fatigue studies published in the scientific literature deal with lead-based materials. Detailed reviews were given by Genenko et al. and by Lupascu and Rödel for bulk PZT.6,7) PZT ceramics show high piezoelectric properties, however, due to toxic by-products lead-free alkali niobate ceramics such as LNKN with several additives at high electric fields and different temperatures to observe the influence of phase transition on this fatigue-free behavior was discussed.8–10)

2. Experimental

Two different materials were used for the fatigue tests: Li0.06Na0.52K0.42NbO3 (LNKN6-a) and Li0.06Na0.52K0.42NbO3 with additives: 0.65 mol % Li2CO3, 1.3 mol % SiO2, 0.2 mol % MnO2, 0.5 mol % SrCO3, and 0.5 mol % ZrO2 (LNKN6-b). More information about the materials can be found in the work by

1) Corresponding author: A. Martin; E-mail: martin.alexander@nitech.ac.jp

The unipolar fatigue behavior of undoped Li0.06Na0.52K0.42NbO3 (LNKN6-a) and the same compound with additives (LNKN6-b) were observed. Unipolar cycling was performed between room temperature and 150°C under a high unipolar electric field of 4 kV/mm. LNKN6-a fatigued under crack formation throughout the examined temperature range. The switchable polarization 2P, of LNKN6-a decreased for 20% (≈2 μC/cm2) at 150°C. In contrast, mechanical degradation was not observed in LNKN6-b. The switchable polarization 2P, of LNKN6-b decreased for 15% (≈3.2 μC/cm2) at 150°C. It should be noted that at 50°C, fatigue-free behavior was observed for LNKN6-b over at least 106 cycles. The influence of phase transition on this fatigue-free behavior was discussed.

Key-words: Actuators, Fatigue, Piezoelectric properties, Lead-free, Phase transformation

©2016 The Ceramic Society of Japan. All rights reserved.

[Received November 10, 2015; Accepted April 3, 2016]
Hatano et al.\textsuperscript{24} The ceramic samples were cut and ground into rectangular bars with a thickness of about 0.6 mm, width of 2.1 mm, and length of 10.5 mm. The largest surfaces were coated with silver and annealed at 700°C for 5 min. Subsequently, the samples were poled in silicone oil under an applied electric field of $E = 3 \text{kV/mm}$ for 30 min and a temperature of 100°C for LNKN6-a and 150°C for LNKN6-b. The samples were cooled down to 40°C with the electric field still switched on. To apply the cyclic electric fields, the samples were placed between a metallic electrode and electrode. To avoid arcing, this set-up was immersed in a bath of silicone oil. A function generator (Multifunction Synthesizer WF 1943A, NF Corporation, Yokohama, Japan) connected to an amplifier (Model 610D, TRex Inc., New York, USA) was used to apply a sinusoidal unipolar signal with a frequency of 50 Hz onto the samples for up to 10\textsuperscript{6} cycles. The amplitude during these fatigue tests was 4 kV/mm; this is approximately four times the coercive field $E_{c}$ for LNKN6-a and approximately 3 times that for LNKN6-b. The fatigue tests were performed at selected temperatures between room temperature and 150°C. Care was taken that the applied electric field was aligned in the same direction as the polarization field of each sample. To obtain reliable results for long-time changes in material characteristics after fatigue tests, 4 samples were tested under each temperature and additional experiments were performed after 48 h. $P(E)$ hysteresis loops were determined with a ferroelectric tester (TF Analyzer 2000, aixAcct, Aachen, Germany) at room temperature. Unipolar strain $S$ was measured via laser Doppler vibrometer (Demodulator AT3600, Graphtec, Yokohama, Japan). The measuring frequency and electric field was 1 kHz and 0.5 kV/mm, respectively.

Silver electrodes were polished off, and X-ray diffraction data were obtained by an X-ray diffractometer (X’Pert Pro, PANalytical, Almelo, Netherlands) with a Cu Kα1 radiation source. Step mode measurements contained step width of 0.017\textdegree and the total spectrum between an angular interval from 10 to 100\textdegree was collected at room temperature. The collected data was refined, using the XRD Rietveld method supported by the software FULLPROF.\textsuperscript{25} We modeled both materials, using space groups P4mm with coexisting Amm2 and used the Pseudo-Voigt function to define the profile shape.

Two samples for each composition and temperature were further used for SEM evaluations. The surfaces vertical to the poling directions of the samples were mechanically polished with SiC powder (#600–#2000) and finished with polishing with colloidal silica. To reveal domain structures, the polished samples were chemically etched for about 45 min with a mixture of HF:HCl:H₂O = 1:1:18 by volume. A thin layer of Au/Pd was sputtered onto the samples to avoid charging; then, domain configurations were observed using a SEM system (JSM-7001F, JEOL, Tokyo, Japan) at an acceleration voltage of 5 kV.

3. Results

The X-ray diffraction (XRD) pattern of LNKN6-a and LNKN6-b are shown in Fig. 1. As shown by the patterns, both samples do not differ much from each other. However, Rietveld refinement showed the difference in phase composition. Table 1 compares the weight percentage wt\% of the tetragonal phase (P4mm) to the orthorhombic phase (Amm2) in both materials before and after poling, as well as different piezoelectric properties.

The Young’s moduli $Y_{11}$ as well as the coupling factor $k_{31}$ for LNKN6-a and LNKN6-b have the same value. Overall, LNKN6-b had a lower weight percentage of the tetragonal phase than LNKN6-b in the poled and unpoled case. For both materials, the weight percentage increased after poling took place. Because $d_{31}$ and $Q_m$ are inversely proportional, LNKN6-a had a higher piezoelectric coefficient $d_{31}$ and lower mechanical quality factor $Q_m$ than LNKN6-b. Figure 2 shows the temperature dependence of the relative permittivity $\varepsilon_{33}/\varepsilon_0$ and the dielectric loss tan $\delta$ for poled LNKN6-a and LNKN6-b.

The dielectric loss tan $\delta$ was on the same level for both materials and increased with increasing temperature. LNKN6-a had higher relative permittivity values ($\varepsilon_r = 900$) at room temperature than LNKN6-b with $\varepsilon_r = 750$. In the observed temperature range,

![Fig. 1. XRD profiles of unpoled LNKN6-a and LNKN6-b.](image_url)

![Fig. 2. Relative permittivity as a function of temperature.](image_url)

Table 1: Weight percentage of tetragonal phase and different piezoelectric properties of LNKN6-a and LNKN6-b. The calculated $R$ values of the Rietveld refinement were about $R_{wp} = 12.7$ and $R_{exp} = 6.9$

| Material       | Condition | Wt\% tetragonal phase (%) | $d_{31}$ (pC/N) | $k_{31}$ (%) | $Q_m$ | $Y_{11} \times 10^{10}$ (N/m²) | Grain size (\textmu m) |
|---------------|-----------|---------------------------|-----------------|--------------|-------|-----------------------------|----------------------|
| LNKN6-a       | unpoled   | 58.5 ± 2.5                | \-               | \-           | \-    | 8.1 ± 0.1                   | 8–10                 |
| LNKN6-a       | as-poled  | 62.0 ± 3.5                | -85 ± 2         | 23.0 ± 0.5   | 50 ± 3 | 8.1 ± 0.1                   | 8–10                 |
| LNKN6-b       | unpoled   | 52.3 ± 2.5                | \-               | \-           | \-    | 8.1 ± 0.1                   | 3–4                  |
| LNKN6-b       | as-poled  | 57.5 ± 3.1                | -60 ± 2         | 22.5 ± 0.2   | 115 ± 4| 8.1 ± 0.1                   | 3–4                  |
the permittivity shows a maximum in both materials around 50°C, with \( \varepsilon_r = 950 \) and \( \varepsilon_r = 800 \) for LNKN6-a and LNKN6-b, respectively. After 50°C, the permittivity of LNKN6-a decreased more significantly than LNKN6-b and showed lower values after a temperature of about 110°C. LNKN6-b exhibited a very stable temperature dependence of the dielectric permittivity over the range of room temperature to 150°C. Thus, the composition of LNKN6-b seems to be closer to the morphotropic phase boundary MPB (tetragonal wt % less than 60%), thereby showing less susceptibility against temperature changes. In addition, the unipolar strain \( S \) was observed for both samples before and after fatigue at different temperatures. The results are shown in Fig. 3, with the solid lines representing the as-poled values of LNKN6-a and LNKN6-b.

The results show that unipolar strain is decreasing for LNKN6-a throughout the observed temperature range. The degradation had equal values for all fatigue temperatures. In contrast, the unipolar strain of LNKN6-b seemed only to decrease at room temperature and remained constant after fatigue between 50 and 150°C. The ferroelectric properties were also changed after fatigue experiments. Figures 4(a)–4(d) represents the \( P(E) \) hysteresis loops of LNKN6-a and LNKN6-b before and after fatigue temperatures of room temperature and 150°C.

In general, LNKN6-b had a higher switchable polarization \( 2P_r \) and coercive field \( E_c \) than LNKN6-a. According to the \( P(E) \) hysteresis loops, the switchable polarization \( 2P_r \) and the coercive field \( E_c \) of unfatigued samples were 10.1 \( \mu \)C/cm\(^2\) and 0.95 kV/mm for LNKN6-a and 21.6 \( \mu \)C/cm\(^2\) and 1.35 kV/mm for LNKN6-b, respectively. In both materials, the switchable polarization \( 2P_r \) decreased after the fatigue experiments. Figure 4(a) shows the fatigue behavior of LNKN6-a under room temperature, whereby an increase of the coercive field \( E_c \) is apparent. In Figs. 4 (b)–4(d), the coercive field \( E_c \) remained constant, and the hysteresis loops shifted. Figures 5(a) and 5(b) show the changes of switchable polarization \( 2P_r \) and \( E_c \) after 10\(^6\) cycles at different temperatures. The values of the fatigued samples \( 2P_r \) and \( E_c \) were normalized by dividing them with the initial values \( 2P_r \) and \( E_c \), respectively. The highlighted line represents the unfatigued state of an as-poled sample.

As mentioned before, the switchable polarization decreased after the fatigue experiments. Figure 5(a) shows the degradation
for LNKN6-a at each temperature condition. Figure 5(b) shows that the switchable polarization decreased for LNKN6-b after each fatigue temperature $T$ except 50°C. On the other hand, the coercive field $E_c$ remained constant for LNKN6-b throughout each condition. For LNKN6-a, the average value of the coercive field after fatigue at temperatures higher than room temperature would also suggest that no fatigue appeared under these conditions. As LNKN6-b remained at the same value as in the unfatigued case, the results (b) LNKN6-b at different fatigue temperatures.

Increasing temperature and reached a minimum at 100°C with a nearly 30% ($=3\mu C/cm^2$) reduction. At 150°C, $2P$, of LNKN6-a and LNKN6-b declined by 20% ($=2\mu C/cm^2$) and 15% ($=3.2 \mu C/cm^2$), respectively. It should be noted that at 50°C, $2P$, of LNKN6-b remained at the same value as in the unfatigued case, suggesting that no fatigue appeared under these conditions. As shown in Figs. 4(a)–4(d), besides changes in coercive field and switchable polarization, the $P(E)$ loops shifted after the fatigue experiments. Due to the poling process, both samples had an internal bias field $E_{bias}$. These residual internal bias fields $E_{bias} = 1/2(|E_c^-| - |E_c^+|)$ were evaluated for both samples to 0.1 and 0.35 kV/mm for LNKN6-a and LNKN6-b, respectively.

Figure 6 shows the internal bias field after the fatigue tests at different fatigue temperatures. The highlighted lines are the values of as-poled LNKN6-a and LNKN6-b.

At lower temperatures, the changes of the internal bias field differ for both materials. After fatigue at room temperature, the internal bias field of LNKN6-a reached negative values of about $-0.1$ kV/mm. At 50 and 100°C the internal bias reached positive values, which were still slightly lower than the poled case. LNKN6-b, showed an increase of the internal bias field at each temperature condition, except 50°C. Only for fatigue tests at 150°C the changes of the internal bias field were similar for both materials. $E_{bias}$ increased for about two times its initial value for LNKN6-a ($=0.29$ kV/mm) as well as for LNKN6-b ($=0.6$ kV/mm). Figure 6 also shows a fatigue-free behavior of LNKN6-b at 50°C. Table 2 summarizes the results of the piezoelectric properties of LNKN6-a and LNKN6-b before and after fatigue at different temperatures. $Y_{11}$ remained constant throughout the experiments for LNKN6-a and LNKN6-b. Furthermore, the piezoelectric properties $k_{31}$ and $d_{31}$ of both materials show an inversely proportional behavior to $Q_m$. For LNKN6-a $k_{31}$ and $d_{31}$ increased under fatigue tests up to temperatures of 100°C, whereas $Q_m$ decreased. At fatigue tests under 150°C, $k_{31}$ and $d_{31}$ decreased and $Q_m$ increased. The piezoelectric properties of LNKN6-b increased at 50°C as well as 100°C. In case of LNKN6-b the $k_{31}$ decreased, whereas the mechanical quality factor $Q_m$ increased.

4. Discussion

After fatigue at several temperatures, no significant changes were observed for piezoelectric properties, but strain $S$ and $P(E)$ hysteresis loops showed different fatigue behaviors depending

![Figure 5](image5.png)

Fig. 5. Normalized $2P$ and coercive field $E_c$ of (a) LNKN6-a and (b) LNKN6-b at different fatigue temperatures.

![Figure 6](image6.png)

Fig. 6. Internal field $E_{bias}$ of fatigued LNKN6-a and LNKN6-b at different fatigue temperatures.

| Material | Physical Quantity | as-poled | 25°C | 50°C | 100°C | 150°C |
|----------|------------------|---------|------|------|-------|-------|
| LNKN6-a  | $d_{31}$ (pC/N)  | $-85 \pm 2$ | $-92 \pm 8$ | $-94 \pm 2$ | $-96 \pm 12$ | $-85 \pm 11$ |
|          | $k_{31}$ (%)     | $23 \pm 0.5$ | $24.1 \pm 1.7$ | $23.9 \pm 0.8$ | $24.8 \pm 1.2$ | $21.8 \pm 0.2$ |
|          | $Q_m$            | $50 \pm 3$ | $45 \pm 1.0$ | $35 \pm 6$ | $39 \pm 14$ | $43 \pm 17$ |
|          | $Y_{11} \times 10^{10}$ (N/m²) | $8.1 \pm 0.05$ | $8.0 \pm 0.3$ | $8.0 \pm 0.3$ | $8.0 \pm 0.4$ | $8.0 \pm 0.7$ |
| LNKN6-b  | $d_{31}$ (pC/N)  | $-60 \pm 2$ | $-56 \pm 0.5$ | $-60 \pm 1$ | $-61 \pm 4$ | $-60 \pm 0.5$ |
|          | $k_{31}$ (%)     | $22.5 \pm 0.2$ | $22 \pm 0.3$ | $22.7 \pm 0.3$ | $22 \pm 0.4$ | $20.9 \pm 0.7$ |
|          | $Q_m$            | $115 \pm 4$ | $129 \pm 8$ | $103 \pm 16$ | $128 \pm 4$ | $135 \pm 7$ |
|          | $Y_{11} \times 10^{10}$ (N/m²) | $8.1 \pm 0.05$ | $8.1 \pm 0.3$ | $8.1 \pm 0.2$ | $8.1 \pm 0.4$ | $8.1 \pm 0.05$ |
on the material and temperature. For LNKN6-a after each fatigue experiment as well as for LNKN6-b at room temperature, the strain was lower compared to as-poled samples. The switchable polarization $2P$ decreased, and an internal bias field $E_{bias}$ was observed. In addition, the coercive field $E_c$ remained relatively constant. Two noteworthy observations were made for the fatigue conditions of LNKN6-a under room temperature and LNKN6-b under 50°C. First, LNKN6-a showed an increased coercive field after fatigue at room temperature. Furthermore, no fatigue was observed for LNKN6-b after tests at 50°C. In many cases, the fatigue behavior was similar to the observations made in the literature for PZT. Fatigue studies on PZT explain different processes that can appear during electric cycling of piezoelectric materials. The inherent processes responsible for unipolar fatigue are attributed to induced charge carrier accumulation during electric field cycling. The domain mobility is not significantly affected by these charge carrier accumulations and it is therefore more likely that these accumulations develop at grain boundaries.30) The main charge carriers in PZT structures are oxygen vacancies31) and thus unipolar fatigue effects can be attributed to their accumulation.14) This accumulation leads to an increase of an internal bias field as well as a decrease of the switchable polarization.51,14,28,30) Yao et al. explained the development of the internal bias field during unipolar fatigue. At grain boundaries the polarization vectors of domains in neighboring grains cannot compensate each other due to crystallographic mismatch. This leads to the occurrence of strong, locally varying depolarization fields. Charge carriers, such as oxygen vacancies, try to compensate these fields and accumulate at the respective areas. This accumulation of charges forms the local bias field $E_{local}$, which has the same orientation as the applied field during fatigue.14) In order to explain the larger bias field in LNKN6-b, we took a closer look at the dopants. From the ion radii of the different elements, we can estimate if they attach to A or B site of the perovskite structure. For this reason Table 3 lists the different dopants with their respective ion radius. It should be noted that the values for B site ions were taken with a coordination number of 6, whereas the A site ions were taken with the highest possible coordination number.31)

| Element | Li$^{+}$ | Sr$^{2+}$ | Mn$^{2+}$/Mn$^{4+}$ | Si$^{4+}$ | Zr$^{4+}$ |
|---------|--------|--------|-----------------|--------|--------|
| Ion-radius (pm) | 92 | 144 | 96/53 | 40 | 72 |

by 2 and 3.2 μC/cm², respectively. Glaum et al. showed the influence of temperature on $E_{bias}$ and observed a higher induced internal bias field during cycling with increasing temperature.30) This behavior was also observed for LNKN6-b, where with increasing temperature from 100 to 150°C, the internal bias field after fatigue increased. However space charge accumulation cannot account for all the observed results. Figures 7(a) and 7(b) show the micro structure of LNKN6-a and LNKN6-b. The line structure visible in both pictures represent the domain structure, which in this case show 90° domain walls and 60° domain walls.

An intragranular crack, visible in Fig. 7(a), is rarely observed during unipolar cycling, because the introduced stresses are usually too small to initiate cracks. Nevertheless, Jiang et al. showed that crack formation and propagation in PZT during unipolar cycling depends on grain size and piezoelectric constant $d_{31}$, porosity32) and surface condition.34) Thus, LNKN6-a is more susceptible to crack formation, due to a higher piezoelectric constant $d_{31}$ and average grain size. After micro-crack formation, the electric field is concentrated at the crack tips and weakened throughout the whole sample. As a result, the effective field inside the sample is less than the applied field, which limits domain switching and consequently decreases polarization and increases the coercive field $E_c$.32) Furthermore, the micro cracks reduced unipolar strain as shown above. In LNKN6-b, no cracks were observed throughout the investigated temperature range, but the decrease in strain at room temperature might suggest the formation of defects in the material. Further, with increasing temperature the crack formation appeared less in LNKN6-a, as suggested by the coercive field measurements from Fig. 5(a) and micro structure observation. Hill et al. showed the same behavior in PZT, where with increasing temperature the crack density after unipolar cycling decreased.31) In case of LNKN6-a, the crack density was the highest after fatigue at room temperature. Because of the negative internal bias field $E_{bias}$ at this temperature, it is suggested that intragranular cracks prevented the material from forming a depolarization field during fatigue. Moreover, LNKN6-a showed larger error bars in Figs. 4 and 5 than LNKN6-b, which can also be the result of micro crack formation, as the process is random. Nevertheless, the fatigue free behavior of LNKN6-b at 50°C cannot be explained by the above mentioned processes. Representative impedance measurements are shown in Figs. 8(a)-8(d) for LNKN6-b at different temperatures. In the figure, the bold lines represent the evaluations of the unfatigued sample, and the dotted lines describe the results of the measurements 48 h after the fatigue experiments. The arrows point in the direction of the changes before and after fatigue tests in the frequencies.

The figures show for LNKN6-b samples the fatigue behavior variation with the applied temperature during the tests. At room
temperature, resonant and antiresonant frequencies shifted to higher values, together with the maximum in the phase angle. At 50°C, no changes were detected and the sample remained free of fatigue. However, above 50°C, fatigue occurred by shifting resonant and antiresonant frequencies slightly to lower values.

For LNKN6-a, the measured results shifted to lower frequencies throughout the investigated temperature range.

Figure 9 shows the changes of the weight percentage of the tetragonal phase, which were evaluated from the Rietveld refinement. The highlighted line represents the tetragonal weight percentage of the as-poled samples.

The change in the weight percentage of tetragonal phase showed that phase transformation partially appeared during fatigue experiments with increasing fatigue temperature. LNKN6-a showed a decrease in the tetragonal phase after fatigue under room temperature and 50°C. At 100 and 150°C, the tetragonal phase in LNKN6-a showed no changes compared to as-poled samples. On the other hand, the tetragonal phase of LNKN6-b remained almost unchanged during fatigue at room temperature and 50°C. At temperatures above 50°C, the material showed an increase of the tetragonal phase with increasing temperature. It is therefore believed, that partial phase transformation from orthorhombic to tetragonal phase occurred during fatigue experiments and that this process counteracted the space charge accumulation. As a result, no fatigue was observed in LNKN6-b at 50°C.

5. Conclusions

The unipolar fatigue behavior of lithium-modified NKN ceramics have been evaluated as a function of temperature in the range from room temperature to 150°C. In general, the piezoelectric properties in both materials did not decrease significantly. However, it was shown that LNKN6-a is especially susceptible to micro crack formation during fatigue at room temperature up to 150°C. These intergranular cracks lead to a significant decrease of the switchable polarization $2P_r$. This behavior was not observed in LNKN6-b, which showed no micro crack formation throughout the examined temperature range. The major factor was most likely the smaller grain size compared to LNKN6-a. Notably in LNKN6-b is the fatigue free behavior at 50°C. It is suggested, that possible phase transformation lead to this behavior in LNKN6-b.
Acknowledgements  This research was supported by a JSPS Grant-in-Aid for Scientific Research (B) and by the Industrial Technology Research Grant Program from the New Energy and Industrial Technology Development (NEDO) of Japan.

References
1) J. E. Huber, N. A. Fleck and M. F. Ashby, Proc. R. Soc. Lond. A, 453, 2185–2205 (1997).
2) M. Zupan, M. F. Ashby and N. Fleck, Adv. Eng. Mater., 4, 933–940 (2002).
3) E. Aksel and J. L. Jones, Sensors (Basel Switzerland), 10, 1935–1954 (2010).
4) N. Balke, D. C. Lupascu, T. Granzow and J. Rödel, J. Am. Ceram. Soc., 90, 1081–1087 (2007).
5) H. Wang, S.-M. Lee, J. L. Wang and H.-T. Lin, J. Appl. Phys., 116, 234101 (2014).
6) Y. A. Genenko, J. Glaum, M. J. Hoffmann and K. Albe, Mater. Sci. Eng., B, 192, 52–82 (2015).
7) D. Lupascu and J. Rödel, Adv. Eng. Mater., 7, 882–898 (2005).
8) Y. Saito, H. Takao, T. Tani, T. Nonoyama, K. Takatori, T. Homma, T. Nagaya and M. Nakamura, Nature, 432, 84–87 (2004).
9) S. Priya and S. Nahm, Lead-free piezoelectrics. New York: Springer, 2012 (2012).
10) F.-Z. Yao, K. Wang, L.-Q. Cheng, X. Zhang, W. Zhang, F. Zhu and J.-F. Li, J. Am. Ceram. Soc., 98, 448–454 (2015).
11) S. Zhang, R. Xia, H. Hao, H. Liu and T. R. Shroult, Appl. Phys. Lett., 92, 152904 (2008).
12) J. Hao, Z. Xu, R. Chu, W. Li and P. Fu, Mater. Res. Bull., 65, 94–102 (2015).
13) E. Patterson and D. Cann, IEEE Trans. Ultrason. Ferroelectr. Freq. Control, 58, 1835–1841 (2011).
14) F. Z. Yao, J. Glaum, K. Wang, W. Jo, J. Rödel and J. F. Li, Appl. Phys. Lett., 103, 192907 (2013).
15) M. S. Senousy, R. K. N. D. Rajapakse, D. Mumford and M. S. Gadala, Smart Mater. Struct., 18, 045008 (2009).
16) G.-L. Zhao, H. Zhang, B.-P. Zhang and J.-F. Li, Ceram. Int., 38, 583–587 (2010).
17) Y. Guo, K. Kakimoto and H. Ohsato, Appl. Phys. Lett., 85, 4121–4123 (2004).
18) M. Matsubara, T. Yamaguchi, K. Kikuta and S. Hirano, Jpn. J. Appl. Phys., 44, 6136–6142 (2005).
19) E. Hollenstein, D. Damjanovic and N. Setter, J. Eur. Ceram. Soc., 27, 4093–4097 (2007).
20) R. E. Jaeger and L. Egerton, J. Am. Ceram. Soc., 45, 209–213 (1962).
21) G. H. Haertling, J. Am. Ceram. Soc., 50, 329–330 (1967).
22) C. W. Ahn, S. Nahm, M. Karmarkar, D. Viehland, D. H. Kang, K. S. Bae and S. Priya, Mater. Lett., 62, 3594–3596 (2008).
23) H. E. Mgbemere, R.-P. Herber and G. A. Schneider, J. Eur. Ceram. Soc., 29, 1729–1733 (2009).
24) K. Hatano, A. Yamamoto, Y. Doshida and Y. Mizuno, J. Ceram. Soc. Japan, 123, 561–564 (2015).
25) T. Roisnel and J. Rodríguez-Cardavjal, Mater. Sci. Forum, 378–381, 118–123 (2001).
26) C. Verdier, D. C. Lupascu and J. Rödel, Appl. Phys. Lett., 81, 2596–2598 (2002).
27) B. Guiffard, E. Boucher, L. Eyraud, L. Lebrun and D. Guyomar, J. Eur. Ceram. Soc., 25, 2487–2490 (2005).
28) Z. Luo, T. Granzow, J. Glaum, W. Jo, J. Rödel and M. Hoffmann, J. Am. Ceram. Soc., 94, 3927–3933 (2011).
29) Z. Luo, J. Glaum, T. Granzow, W. Jo, R. Dittmer, M. Hoffmann and J. Rödel, J. Am. Ceram. Soc., 94, 529–535 (2011).
30) J. Glaum, T. Granzow, L. A. Schmitt, H.-J. Kleebe and J. Rödel, Acta Mater., 59, 6083–6092 (2011).
31) R. D. Shannon, Acta Crystallogr., Sect. A: Cryst. Phys., Diffr., Theor. Gen. Crystallogr., 32, 751–767 (1976).
32) Q. Jiang, E. C. Subbarao and L. E. Cross, Acta Metall. Mater., 42, 3687–3694 (1994).
33) Q. Y. Jiang and L. E. Cross, J. Mater. Sci., 28, 4536–4543 (1993).
34) Q. Jiang, W. Cao and L. E. Cross, J. Am. Ceram. Soc., 77, 211–215 (1994).
35) M. D. Hill, G. S. White, C.-S. Hwang and I. K. Lloyd, J. Am. Ceram. Soc., 79, 1915–1920 (1996).

736