Reduction effects of orientation control on nonlinear piezoelectricity in (K, Na)NbO$_3$ system ceramics

Keisuke ISHII* and Shinjiro TASHIRO

Department of Materials Science and Engineering, The National Defense Academy,
1–10–20 Hashirimizu, Yokosuka, Kanagawa 239–8686, Japan

The influence of crystalline grain orientation on nonlinear piezoelectricity in lead-free (K, Na)NbO$_3$ system ceramics was studied. The orientation control was accomplished using a rolling-extension method with plate-like NaNbO$_3$ single-crystal particle templates prepared by single-step molten salt synthesis. A negative correlation between the orientation factor and the nonlinear piezoelectricity was observed. The nonlinear coefficient $\varepsilon_{33}$ measured in textured samples with an orientation factor of 97% decreased to one-sixth that of randomly oriented samples.

Key-words : Piezoelectric ceramics, Alkali niobate-based ceramic, Grain orientation control, Nonlinear piezoelectricity, Current jump

1. Introduction

As an inevitable result of the downsizing and increasing driving power of piezoelectric ceramic devices for high-power application, the driving power density of these devices has rapidly increased in recent years. Consequently, the appearance of nonlinear piezoelectric phenomena, such as the generation of higher harmonic voltages, jump phenomena of sample current or of piezoelectric strain, changes in resonance frequency, and increased electrical loss associated with rapid heat generation, has come to stand out. These phenomena interfere with further increases in the driving-power density of the devices. In order to suppress the nonlinear piezoelectricity, acceptors have been added to materials. Furthermore, although the suppression mechanisms are not yet clear, various additives of the third component materials have also been employed on the basis of empirical knowledge. These methods are a kind of material composition optimization, and have been reported to decrease the nonlinearity to approximately 20%. However, even when acceptors are added to them, (K, Na)-NbO$_3$(KNN) ceramics, the leading alternative candidate to lead-based ceramics have a simple perovskite structure, which has poor anisotropy in the crystal structure and growth, effective methods of grain orientation are limited. In the present study, NaNbO$_3$-NN single-crystal particles with plate-like shapes prepared by a single-step molten salt synthesis developed by the authors, were employed for grain orientation control. Using these high-aspect-ratio NN particles as templates, grain orientation control was performed for KNN ceramics by employing the rolling-extension orientation method, which was also developed by the authors. The rolling-extension orientation technique is a reactive template grain growth method, which can easily fabricate highly textured ceramics with a large rectangular shape that would be difficult to prepare by conventional grain orientation techniques.

Grain orientation techniques are methods of arranging the crystallographic orientation of the crystalline grains in a ceramic, and have mainly been developed to improve linear piezoelectricity. On the other hand, it has been reported that the orientation of rod-shape crystal grains suppresses the change in resonance frequency and the appearance of the current-jump phenomenon in tungsten-bronze structured ferroelectric ceramics. This report suggests the possibility that grain orientation techniques reduce the nonlinear piezoelectricity. However, the relationship between nonlinear piezoelectricity and grain orientation has not been reported for lead-free piezoelectric ceramics with high practicality, such as KNN system ceramics. This is partially due to the difficulty of fabricating KNN system ceramic samples with the high orientation factor, rectangular shape, and large size required for a quantitative estimation of the nonlinear piezoelectricity.

In conventional grain orientation techniques, crystal growth anisotropy or magnetic anisotropy of the crystals is frequently employed. On the other hand, since KNN system ceramics have a simple perovskite structure, which has poor anisotropy in the crystal structure and growth, effective methods of grain orientation are limited. In the present study, NaNbO$_3$-NN single-crystal particles with plate-like shapes prepared by a single-step molten salt synthesis developed by the authors, were employed for grain orientation control. Using these high-aspect-ratio NN particles as templates, grain orientation control was performed for KNN ceramics by employing the rolling-extension orientation method, which was also developed by the authors. The rolling-extension orientation technique is a reactive template grain growth method, which can easily fabricate highly textured ceramics with a large rectangular shape that would be difficult to prepare by conventional grain orientation techniques.

In this study, by comparing the nonlinear piezoelectricity of conventional randomly oriented samples with that of textured samples with various orientation factors, the effects of grain orientation on the nonlinear piezoelectricity were studied for KNN ceramics. In order to quantitatively estimate the non-

* Corresponding author: K. Ishii; E-mail: kishii@nda.ac.jp

DOI http://dx.doi.org/10.2109/jcersj2.123.114

©2015 The Ceramic Society of Japan. All rights reserved.
linear piezoelectricity, a nonlinear coefficient for the third-order term of the electric field $\varepsilon_D^{(3)}$ was used. $\varepsilon_D^{(3)}$ was calculated from the third-order harmonic voltages generated in the samples 3,5,10,12,18,22-24.

2. Sample preparation and estimation method

$(K_{0.5}Na_{0.5})NbO_3$ calcined powder for fabricating textured and randomly oriented samples was prepared by a conventional solid-phase process. As starting materials, stoichiometric amounts of K$_2$CO$_3$, Na$_2$CO$_3$, and Nb$_2$O$_5$ reagents were mixed, 1 at % CuO was added as a sintering aid and acceptor, and then the mixture was ground in ethanol for 12 h. The mixture was calcined at 800°C for 5 h and then ground in ethanol for 24 h.

Contrary to the conventional two-step molten salt synthesis, 27,33 both reactions, the preparation of Bi$_{2.5}$Na$_{3.5}$Nb$_5$O$_{18}$ (BNNN5) crystal particles of bismuth-layer structured ferroelectrics and the conversion of plate-like BiNN5 to NN particles by removing bismuth, were simultaneously promoted in the single-step molten salt synthesis of the plate-like single crystal NN particles, hereinafter referred to as NN templates. Single-crystal particles with smooth surfaces, which are desirable for a template, were prepared by this synthesis, whereas polycrystalline particles with rugged plate surfaces are obtained from a two-step molten salt synthesis. 27,40 The fabrication process was described in detail in a previous report. 32,40,41 Bi$_2$O$_3$, Na$_2$CO$_3$, and Nb$_2$O$_5$ reagents as starting materials were mixed in a molar ratio of 5:14:20 in ethanol for 12 h. After drying, NaCl was added to the mixture at a 3:1 NaCl to mixture weight ratio, and mixed with a mechanically driven mortar and pestle for 30 min. After heating in a Pt crucible at 1225°C for 12 h, the products were washed in deionized hot water several times, and sieved with a #250 mesh to remove by-products. The fabricated plate-like NN particles were single crystals with an aspect ratio higher than ten, as shown in the SEM image of Fig. 1.

The NN templates were mixed with the KNN calcined powder (matrix), an organic binder, and an organic solvent, and the mixture was damp-dried and kneaded until it reached claylike softness. The ratio of the weight of the NN templates to the total weight of the templates and the matrix was 5%. 6-mm-thick claylike green sheets were sandwiched between separating films and extended with a roller, as shown in Fig. 2. In order to prepare textured samples with various orientation factors, the thickness extension was done to different extension ratios (the thickness ratio of the green sheets before and after extension) and with different numbers of the extensions. The extension conditions are summarized in Table 2. The extended green sheets for T3 and T4 were folded to a thickness of 6-8 mm and extended again. The finally extended green sheets were stamped into rectangular bars, shape formed and dewaxed at 400°C for 4 h, and then fired using a dual-step sintering process, in which the firing conditions of the first and second steps were 1000°C for 2 h and 1180°C for 12 h, respectively. 32 Textured rectangular samples with standard sizes of 25–27 x 4 x 2 mm$^3$ were fabricated, as illustrated in Fig. 3(a).

Randomly oriented KNN ceramics with a standard size of 30 x 4 x 1 mm$^3$ were also fired by dual-step sintering, in which the firing conditions of the first and second steps were 920°C for 8 h and 1050°C for 2 h, respectively.

The crystal phase was examined by X-ray diffraction (XRD) analysis (Rigaku MiniFlex), and the configuration of the NN templates was observed with field-emission scanning electron microscopy (FE-SEM; Hitachi S-4500). The orientation factor $F$ for the pseudo-tetragonal (100)(001) faces perpendicular to the poling direction was determined by the Lotgering method using
in the thickness direction, and the mechanical strain in the transverse direction, respectively, as shown in Fig. 3(a).

Since only the third nonlinear term in Eq. (2) induces typical nonlinear piezoelectric phenomena such as current-jumping or a change of resonance frequency,9,10,11,12,13,19 the second nonlinear term is negligible. Although the nonlinear piezoelectric coefficient for stress and the imaginary component of $\xi_{D31}$ are required for a more rigorous analysis, only the real component of $\xi_{D31}$ was measured in this study to simplify the analysis. When the sample is driven at close to the fundamental resonance frequency, the circuit equation, Eq. (3), can be obtained from Eq. (2).3)

$$v_3 = L_1 \frac{di_3}{dt} + R_i i_3 + \frac{1}{C_1} \int i_3 dt + \xi_{D31}' \left( \int i_3 dt \right)^3$$  

(3)

Here, $L_1$, $C_1$, and $R_i$ are the motional impedance parameters of the equivalent circuit represented in Fig. 3(b), and $v_3$, $i_3$, and $\omega$ are the sample voltage between the electrodes, the driving current, and the driving angular frequency. The relationship between the material constant $\xi_{D31}$ and the effective nonlinear coefficient for a rectangular vibrator $\xi_{D31}'$ is expressed in the following equation.3,43)

$$\xi_{D31}' = \frac{nb}{A} \xi_{D31}$$  

(4)

Here, $A$ and $b$ are the electrode area and sample thickness, respectively, and $n$ is the conversion factor calculated with consideration of the electrode configuration, which is equal to $3\pi^2/128$ when the rectangular samples have whole-surface electrodes.3,43)

When a sample is driven by a current $i = I_0 \sin \omega t$, the fundamental frequency component of $v$ obtained from Eq. (3), $V_0 \sin(\omega t + \theta)$, is expressed as the following equations.

$$V_0 \sin(\omega t + \theta) = \left( \omega L_1 - \frac{1}{\omega C_1} \right) I_0 \cos \omega t + R_i I_0 \sin \omega t - \frac{3 \xi_{D31}' I_0^3 \cos \omega t}{4So^3}$$

$$= \sqrt{ \left( \left( \omega L_1 - \frac{1}{\omega C_1} \right) I_0 - \frac{3 \xi_{D31}' I_0^3}{4So^3} \right)^2 + (R_i I_0^2 \sin(\omega t + \theta))^2}$$

where,

$$\theta = \tan^{-1} \left( \frac{\omega L_1 - \frac{1}{\omega C_1} - \frac{3 \xi_{D31}' I_0^2}{4So^3}}{R_i I_0^2} \right)$$  

(5)

The third term on the right-hand side of Eq. (5) is a nonlinear voltage of the fundamental frequency component. Furthermore, another nonlinear voltage of the third-order harmonic component is introduced from Eq. (3) adding to the fundamental frequency component of $v$ estimated by measuring this third-order harmonic voltage generated in the samples during constant-current driving.3,5,7,10,12,18) The driving angular frequency is the fundamental resonance angular frequency of the length-extensional $1/2 \lambda$ mode, $\omega_0$. The waveforms of the sample current and sample voltage were simultaneously accumulated into a digital storage scope (Lecroy, 6050-I) in the constant-current-measurement circuit, as shown in Fig. 4(a).3,5)

Using the equivalent circuit of the samples shown in Fig. 3(b), the amplitude of the third-harmonic voltage $V_{3\lambda}$, which is generated in the samples driven by constant current $i = I_0 \sin \omega_0 t$, and that of the apparent voltage observed between the sample electrodes $V_{3\lambda}'$ are expressed as Eq. (6), which was used to calculate $\xi_{D31}'$.3,5)

$$V_{3\lambda} = \frac{1}{\omega_0 C_1} \frac{1}{2 \lambda} I_0 \left( \frac{3 \xi_{D31}' I_0^3}{4So^3} \right)$$

$$V_{3\lambda}' = \frac{1}{2 \lambda} \frac{1}{\omega_0 C_1} \frac{1}{2 \lambda} I_0 \left( \frac{3 \xi_{D31}' I_0^3}{4So^3} \right)$$

(6)

3. Measurement of nonlinear piezoelectricity

We analyzed the nonlinear piezoelectricity using third-order nonlinear piezoelectric equations transformed into the $h$-form equations from the $g$-form, which are expressed as Eq. (2).3,5,7,10,12,18)

$$E_3 = -\frac{g_{31}}{s_{11}} D_1 + \beta_3 S_3 D_3 + \gamma_{D31} D_3^2 + \xi_{D31} D_3^3$$  

(2)

Here, $\xi_{D31}$ and $\gamma_{D31}$ are the intrinsic nonlinear coefficients of the material constants of the second- and third-order terms for the electric field, respectively. $g_{31}$, $\beta_3$ and $s_{11}$ are the piezoelectric $g$ constant, inverse permittivity, and elastic compliance, respectively. $E_3$, $S_3$, and $D_3$ are the electric field, the electric flux density and the mechanical strain in the transverse direction, respectively. The mechanical strain in the transverse direction, respectively, is shown in Fig. 3(a).
where, 

\[ Z_{m3} = R_3 + j \left( 3L_3\omega_0 - \frac{1}{3C_3\omega_0} \right), \quad Z_{Cs3} = \frac{1}{j3\omega_0C_3} \]  

(6)

Here, \( L_3, C_3, \) and \( R_3 \) are the motional impedance parameters for the third resonance, and \( Z_{m3} \) and \( Z_{Cs3} \) are the motional impedance for the third resonance and the impedance due to the damping capacitance at \( \omega_0 \), respectively. Since high-power driving induces a slight change in the elastic compliance of the samples, \( \xi_{D31}' \) was calculated using an equation in which the change in the elastic compliance was compensated.\(^{10}\) Subsequently, \( \xi_{D31}' \) was converted to \( \xi_{D31} \) using Eq. (4).\(^{12}\)

For the measurement of the current-jumping phenomena, the samples were driven at a constant voltage sinusoidal waveform using a constant-voltage circuit, as shown in Fig. 4(b). After amplification of the AC signal detected with a current probe (Tectronix, P6022), the signal was transformed into a DC signal with a full-wave rectifier circuit, and the DC signal was stored in a digital storage scope. The current-jumping phenomena were measured by changing the driving frequency with a sweep rate of 1 kHz/s.

4. Results and discussion

Figure 5 shows XRD profiles measured at the upper surface of each KNN sample. For easy comparison, the values along the vertical axis in each profile were multiplied by the respective proper integral factors. While the intensity of the (110)(011) peak was highest in the randomly oriented sample, the relative intensity of the (800)(000) peaks was higher in the textured samples. The relative intensity increased with increasing extension ratio in the rolling-extension process. Figure 6 shows the (800)(000) orientation factor \( F \), calculated using Eq. (1) and the measurement results shown in Fig. 5. With increasing extension ratio and repetition number, the orientation factors improved, exceeding 95% in the T4 sample fabricated by repeating the orientation process twice to an extension ratio of six. The plate-like NN templates rotate under the shearing force of the rolling-extension, and the top surfaces, or (000)(110) faces, are oriented parallel to the direction of extension. Since the shearing force is enhanced with increasing extension ratio, the orientation degree of the NN templates is improved. It is further improved by repetition of the rolling-extension process. This orientation of the templates in the green sheets increases the orientation factor of the textured sintered samples.

Figure 7(a) shows the effects of the orientation factor on \( d_{33} \) and \( d_{31} \). Both the linear piezoelectric constants increased with increasing orientation factor, and the values for T4 with an orientation factor of 97% became almost twice as high as for R. These results confirmed that the linear piezoelectricity improved with increasing orientation factor. On the other hand, mechanical quality factor \( Q_m \) decreased with increasing orientation factor as shown in Fig. 7(b). The similar tendency in KNN system ceramics has been observed by others.\(^{44}\)

Figure 8 shows the voltage waveforms of R and T4 samples driven at the fundamental resonance frequency \( \omega_0 \) with a current density amplitude of 15 mA/cm\(^2\) measured using the constant-current circuit shown in Fig. 4(a). The waveform of T4 has an almost sinusoidal shape which is similar to the voltage waveform provided by the power supply. On the other hand, the waveform
measured in R is obviously distorted, and large higher-harmonic voltages were generated in the samples. The amplitude of the apparent third-harmonic voltage between the electrodes \( V_{h3} \) was computed from the waveform using a fast Fourier transform, and the effective nonlinear coefficient for the rectangular vibrator \( D_{31}^{\text{B}} \) and the intrinsic nonlinear piezoelectric coefficient as a material constant \( D_{31} \) were calculated subsequently. For a quantitative comparison of the nonlinear piezoelectricity specific to the material the material constant \( D_{31} \) is needed. The relationship between the orientation factor and these nonlinear piezoelectric coefficients obtained for textured and randomly oriented samples is shown in Fig. 9. The absolute value of \( D_{31} \) in the randomly oriented sample is much larger than the value observed in the conventional lead-based piezoelectric samples (\( \leq 3 \times 10^{13} \) Vm²/C²) as shown in Table 1. A similar tendency has been reported by others.\(^ {26} \) The absolute values of \( D_{31} \) decreased with increasing orientation factor. The absolute value for T4 with an orientation factor of 97\% decreased to one-sixth that of R, and was nearly equal to the maximum value measured in the lead-based samples. Figure 10 illustrates the relationship between the resonance frequency and the sample current measured by the constant-
current circuit shown in Fig. 4(a). \( f_0 \) and \( f_0' \) are the resonance frequencies measured with a small signal and under high-power driving conditions, respectively. \( f_0' \) decreased with increasing sample current. The amount of decrease observed in \( R \) was larger than in \( T4 \). From Eq. (5), the \( f_0' \) value at which the phase difference between the sample current and voltage becomes zero (\( \theta = 0 \)) is expressed by the following equation.

\[
2\pi f_0' = \omega_0' = \frac{1 + \sqrt{1 + 3E'_\text{C} \xi_{D31}^2 f_0^2}}{2L C_1}
\]

Since the above equation indicates that the magnitude of the \( f_0' \) change is positively correlated with the absolute value of \( \xi_{D31} \), the experimental results shown in Fig. 10 are consistent with those shown in Fig. 9. Here, the decrease in \( f_0' \) with increasing sample current indicates that the sign of \( \xi_{D31} \) is negative, similar to that of lead-based piezoelectric ceramics.13-12

Figure 11 shows current-frequency spectra around \( f_0 \) in R and T4 samples, measured using the constant-voltage circuit shown in Fig. 4(b). The open circles show currents measured when the driving frequency was swept from lower frequencies to higher frequencies, and the hatched circles show values measured when the frequency was swept from higher to lower values. The shapes of the current changes are asymmetric with respect to a vertical line at \( f_0 \), and the frequencies at which the maximum current was measured were lower than \( f_0 \). This asymmetry is more prominent for R samples. Moreover, obvious hysteresis appeared in the spectra of R. These are typical nonlinear-piezoelectric phenomena called current-jumping, and are also known as “soft-spring” effects in the mechanical nonlinear vibrations.14,45 From a phenomenological viewpoint, these phenomena are considered to be due to softening of the samples during high-vibration-velocity driving.14,45 The sample softening means the apparatus increase of \( f_0 \) is low in the MPB samples.5,8,22,25 As shown in Fig. 7(b) and Fig. 9, \( \xi_{D31} \) and \( Q_\text{m} \) decrease with increasing orientation factor. This is similar to the tendency observed in lead-based samples near the MPB compositions. It seems that the nonlinear movement of the non-180° residual domain walls is suppressed in the samples with the high orientation factor because of the reduction of the localized interior stress.

Since the growth direction for each crystal grain is random in the randomly oriented samples, the spontaneous polarization axis of each domain is different from the applied direction of the poling voltage. Hence, the non-180° residual domains grow in quantity in randomly oriented samples. In addition, the strains after poling are non-uniform among the crystal grains in the randomly oriented samples. This non-uniformity will generate localized interior stresses and strains around the grain boundaries, which inhibit the reversible linear vibration of the domain walls during the driving. The non-180° residual domain walls and their nonlinear behavior are reduced when the crystal grains are arranged, because the orientations of the spontaneous polarization axes are close to the poling direction. This is thought to cause the reduction of nonlinearity with grain orientation control.

5. Conclusions

This work confirms for the first time that grain orientation control can be used to reduce the nonlinear piezoelectricity in KNN system piezoelectric ceramics.

(1) The nonlinear piezoelectric coefficient \( \xi_{D31} \) of the material constant decreases with increasing orientation factor. Compared with randomly oriented samples, \( \xi_{D31} \) in textured samples with an orientation factor of 97% diminishes by a factor of six.

(2) The generation of the third order harmonic voltage, the change in resonance frequency, and the appearance of the current-jumping phenomena are suppressed in textured samples.

References

1) D. Guyomar, N. Aurelle and L. Eyraud, J. Phys. Ill France, 7, 1197–1201 (1997).

2) N. Aurelle, D. Guyomar, C. Richard, P. Gonnard and L. Eyraud, Ultrasonic, 34, 187–191 (1996).

3) S. Tashiro, K. Ishii and K. Nagata, J. Ceram. Soc. Japan, 110, 649–655 (2002).

4) K. Ishii, N. Akimoto, S. Tashiro and H. Igarashi, J. Ceram. Soc. Japan, 106, 555–558 (1998).

5) S. Tashiro, W. Tokunaga, K. Ishii and K. Nagata, Jpn. J. Appl. Phys., 44, 7032–7036 (2005).

6) S. Tashiro, W. Tokunaga, K. Ishii and K. Nagata, Jpn. J. Appl. Phys., 44, 7032–7036 (2005).

7) S. Tashiro, K. Ishii and K. Nagata, Jpn. J. Appl. Phys., 42, 6068–6073 (2003).

8) K. Ishii and S. Tashiro, J. Ceram. Soc. Japan, 116, 1214–1221 (2008).

9) K. Ishii and S. Tashiro, J. Ceram. Soc. Japan, 119, 29–34 (2011).

10) K. Ishii, S. Tashiro and K. Nagata, J. Ceram. Soc. Japan, 111, 401–406 (2003).

11) H. Shimizu, Y. Doshiba, Y. Mizuno, S. Tanaka, K. Uematsu and H. Tamura, Jpn. J. Appl. Phys., 51, 09LD02 (2012).

12) K. Ishii and S. Tashiro, Jpn. J. Appl. Phys., 46, 7038–7052 (2007).

13) T. Wada, IEEE Trans., 119-A, 246–253 (1999) [in Japanese].

14) “Piezoelectric Ceramics and its Applications”, Denpa-Shinbun Co., Tokyo (1974) pp. 38–41 [in Japanese].

15) Y. Nakagawa, A. Saito and T. Maeno, Trans. Jpn. Soc. Mech. Eng. C, 72, 708–715 (2007) [in Japanese].
16) K. Ishii, N. Akimoto, S. Tashiro and H. Igarashi, *J. Eur. Ceram. Soc.*, 19, 1157–1160 (1999).
17) K. Ishii, T. Yamada, S. Tashiro and H. Igarashi, *Trans. Mater. Res. Soc. Jpn.*, 24, 3–26 (1999).
18) K. Ishii, S. Tashiro and K. Nagata, *Trans. Mater. Res. Soc. Jpn.*, 27, 265–268 (2002).
19) O. Ise, K. Satoh and Y. Mamiya, *Jpn. J. Appl. Phys.*, 38, 5531–5534 (1999).
20) Y. Cho and F. Fumihiro, *J. IEICE*, J76-A, 153–160 (1993) [in Japanese].
21) S. Hirose and S. Takahashi, *Jpn. J. Appl. Phys.*, 41, 09MD01 (2012).
22) K. Ishii, T. Murata, S. Tashiro and H. Igarashi, *Jpn. J. Appl. Phys.*, 39, 5597–5599 (2000).
23) K. Ishii, T. Murata, S. Tashiro and H. Igarashi, *ferroelectrics*, 262, 107–112 (2001).
24) S. Tashiro, T. Murata, K. Ishii and H. Igarashi, *Jpn. J. Appl. Phys.*, 40, 5679–5682 (2001).
25) K. Ishii and S. Tashiro, *Jpn. J. Appl. Phys.*, 49, 09MD01 (2010).
26) M. Hagiwara, S. Takahasi, T. Hoshina, H. Takeda and T. Tsurumi, *IEEE Trans. Ultrason. Ferroelectr. Freq. Control*, 58, 1721–1729 (2011).
27) Y. Saito, H. Takao, T. Tani, T. Nonoyama, K. Takatori, T. Homma, T. Nagaya and M. Nakamura, *Nature*, 432, 84–87 (2004).
28) K. Kakimoto, *Bull. Ceram. Soc. Jpn.*, 47, 469–473 (2012) [in Japanese].
29) K. Wang and J.-F. Li, *J. Adv. Ceram.*, 1, 24–37 (2012).
30) K. Kakimoto, K. Ando and H. Ohsato, *J. Eur. Ceram. Soc.*, 30, 295–299 (2010).
31) Z.-Y. Shen, Y. Xu and J.-F. Li, *Ceram. Int.*, 38S, S331–S334 (2012).
32) K. Ishii and S. Tashiro, *Jpn. J. Appl. Phys.*, 52, 09KD04 (2013).
33) A. Kikuchihara and T. Kimura, Proc. 2012 Joint 21st IEEE ISAF/11th IEEE ECAP/IEEE PFM (ISAF/ECAP/PFM), Aveiro, Portugal, (2012) pp. 4–7.
34) T. Tani and T. Kimura, *Adv. Appl. Ceramics*, 105, 55–62 (2005).
35) T. Kimura, *J. Ceram. Soc. Japan*, 114, 15–25 (2006).
36) T. Kimura and T. Tani, *Mater. Integ.*, 17, 16–24 (2004) [in Japanese].
37) S. Tanaka and K. Uematsu, *Bull. Ceram. Soc. Jpn.*, 47, 253–257 (2012) [in Japanese].
38) K. Ishii and S. Tashiro, *Jpn. J. Appl. Phys.*, 116, 1304–1308 (2008).
39) K. Ishii, H. Hashizume and S. Tashiro, *Jpn. J. Appl. Phys.*, 51, 09LC02 (2012).
40) A. Kikuchihara, F. Sakurai and T. Kimura, *J. Am. Ceram. Soc.*, 95, 1556–1562 (2012).
41) K. Ishii, A. Kikuchihara and T. Kimura, *J. Am. Ceram. Soc.*, 96, 2382–2386 (2012).
42) F. K. Lotgering and J. Inorg, *Nucl. Chem.*, 9, 113–123 (1959).
43) K. Ishii, S. Tashiro and K. Nagata, *Jpn. J. Appl. Phys.*, 41, 7095–7098 (2002).
44) H. Takao, Y. Saito, Y. Aoki and K. Horibuchi, *J. Am. Ceram. Soc.*, 89, 1951–1956 (2006).
45) For example, J. Katoh and E. Marui, *Mechanical Vibrational Science*, Corona Co., Tokyo (1972) [in Japanese].
46) D. A. Hall, *J. Mater. Sci.*, 36, 4575–4601 (2001).
47) V. D. Kugel and L. E. Cross, *J. Appl. Phys.*, 84, 2815–2830 (1998).
48) D. Damjanovic, *J. Appl. Phys.*, 82, 1788–1797 (1996).