Application of (K,Na)NbO₃-based lead-free piezoelectric ceramics to ultrasonic sensors

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Ultrasonic sensors fabricated using (K,Na)NbO₃–KTiNbO₅ (KNN–NTK)-based lead-free piezoelectric ceramics were fabricated, their sensor characteristics were compared with those of sensors fabricated using Pb(Zr,Ti)O₃ (PZT)-based piezoelectric ceramics, and effects of material properties on sensor performance were investigated. The sound pressure, sensitivity, temperature increase during driving, and the responsiveness of the KNN–NTK-based sensor equaled those of the PZT-5A-based sensor. PZT-5A is generally used for ultrasonic sensors and has the same mechanical quality factor $Q_m$ as the KNN–NTK ceramics. Most sensor characteristics are explained based on the electromechanical coupling factor and $Q_m$. However, the change in frequency characteristics under high applied power was observed that is attributed to the tendency of the nonlinear phenomenon of KNN–NTK-based ceramics to be weaker than that of PZT-based ceramics.

Key-words: Lead-free piezoelectric ceramics, KNN, Actuator, Sensor, Ultrasonic sensor

1. Introduction

(K,Na)NbO₃ (KNN) Pb-free piezoelectric ceramics are environmentally friendly and have high piezoelectric properties and Curie temperatures. They are therefore expected to find practical applications as alternatives to Pb(Zr,Ti)O₃ (PZT)-based piezoelectric ceramics. KNN-based ceramics in the form of thin films have already been extensively studied, including in device applications such as sensors and microelectromechanical systems. However, for bulk shapes used in many high-power devices, fabricating components that can withstand polarization and driving is difficult because of the easy formation of structural defects such as voids. Thus, the literature contains few examples of KNN-based devices.

We have developed (K,Na)NbO₃–KTiNbO₅ (KNN–NTK) composite Pb-free piezoelectric ceramics composed of KNN, NTK as a second phase, and additional trace elements. The KNN–NTK composites have a high piezoelectric property, a high-density, and a defect-free structure. The manufacturing process of KNN–NTK-based ceramic powders was confirmed to be able to be scaled to 100 kg per batch using spray-drying. Moreover, the application of KNN–NTK ceramics in various devices such as the piezoelectric buzzer and knock sensor are under investigation. Table 1 shows the piezoelectric characteristics of LFP0202 based on JEITA EM-4501A, which is a KNN–NTK-based ceramic, and typical PZT-based ceramics, where $\varepsilon$ is the dielectric constant, $k$ is the coupling factor, $d$ is the piezoelectric strain constant, $g$ is the piezoelectric voltage constant, $Q_m$ is the mechanical quality factor, $\tan \delta$ is the dielectric loss, $T_c$ is the Curie temperature, $Y^\ast$ is the Young’s modulus, $\rho$ is the density, and $\kappa$ is the thermal conductivity.

In previous work, we confirmed that a knock sensor and buzzer fabricated using KNN–NTK-based ceramics exhibited performances equivalent to that of devices fabricated using PZT-based ceramics when the device structures were optimized. However, Table 1 shows that KNN–NTK-based ceramics differed from PZT-based ceramics with respect to mechanical properties such as Young’s modulus, density, and thermal conductivity. Generally, in a piezoelectric device fabricated using PZT-based ceramics, the type of ceramics used as the piezoelectric element is selected based on the coupling constant and $Q_m$ depending on the application. Although the $Q_m$ can well explain vibration properties, it is evaluated through a combination of mechanical material properties such as density and hardness. Hence, in the case of a comparison under conditions where the material systems differ, such as KNN-based and PZT-based systems, the possibility of novel characteristics not being observable with PZT-based systems exists. For example, compared with PZT-based ceramics, LFP0202 has a higher Young’s modulus, lower density, and a higher thermal conductivity.

In the present paper, we fabricated an airborne ultrasonic sensor using a KNN–NTK-based ceramic and...
compared its performance with that of a device fabricated using a PZT-based ceramic; we then discussed the effect of the different material systems on the sensor characteristics. The ultrasonic sensors are used for various applications such as obstacle detection, distance measurement, and parametric speakers. Therefore, good performance traits such as good transmission/reception characteristics, high-power characteristics, and responsiveness are required. In general, soft-PZT, such as PZT-5A, is used as piezoelectric ceramics. Specifically, in the present work, sound pressure, sensitivity, responsiveness, and the temperature increase during driving were evaluated as sensor characteristics; the devices’ behaviors are discussed on the basis of the difference in piezoelectric properties and mechanical properties between the KNN–NTK and PZT piezoelectric ceramics.

2. Experiment

Open-structure airborne ultrasonic sensors (Fig. 1) were fabricated using LFP0202, PZT-5A, and PZT-4 (NGK Spark Plug Co., Ltd.). All components, including the piezoelectric elements used for making the ultrasonic sensors, had the same dimensions. The device structure is similar to conventional ultrasonic sensors.\(^1\) Note that the individual differences in sensors attributed to device assembly have no significance in this discussion; hence, this discussion has been excluded herein.

Piezoelectric properties were measured using a Hewlett-Packard HP 4194A impedance analyzer. Transmission characteristics were evaluated by sound-pressure level (SPL), and reception characteristics were evaluated by sensitivity. SPL was evaluated from sound pressure measured with a standard microphone (46DD, G.R.A.S.S.) installed 30 cm in front of the ultrasonic sensor (0 dB = 20 \mu Pa). The reception characteristics were evaluated from the voltage obtained from the ultrasonic sensor positioned 30 cm from the sound source and from the sound pressure measured with a standard microphone placed alongside the ultrasonic sensor (0 dB = 10 \text{V}_{\text{ref}} \text{ Pa}^{-1}).

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### Table 1. Piezoelectric and mechanical properties of a KNN–NTK based ceramic and typical piezoelectric ceramics

| Base material | Type   | \(\varepsilon_{33}/\varepsilon_{0}\) | \(k_p\) | \(N_p\) /kHz | \(d_{31}\) /pC N m^{-1} | \(g_{11}\) /mV m N^{-1} | \(Q_m\) | \(\tan \delta\) | \(T_r\)/°C | \(Y^f\)/GPa | \(\rho\) /g cm^{-3} | \(k_x\) /W m^{-1} K^{-1} |
|---------------|--------|-----------------|-------|--------------|----------------|-----------------|------|---------------|--------|---------|----------------|------------------|
| KNN–NTK       | LFP0202| 1.400           | 0.49  | 3200         | -105           | 8.3             | 51   | 0.019         | 290    | 89      | 4.5            | 2.4              |
| PZT           | PZT-5A | 1.954           | 0.35  | 2051         | -167           | 10.4            | 60   | 0.020         | 320    | 65      | 7.7            | 1.1              |
| PZT          | PZT-4² | 1.268           | 0.60  | 2293         | -96            | 8.5             | 1350 | 0.004         | 374    | 81      | 7.7            | 1.1              |

\(^1\)PZT-M, \(^2\)PZT-C product in NGK SPARK PLUG CO., LTD.

### Table 2. The driving voltage of ultrasonic sensors

| Material       | Driving voltage/V |
|----------------|-------------------|
| LFP0202        | 6.17              |
| PZT-5A         | 3.83              |
| PZT-4          | 4.36              |

### Table 3. Static piezoelectric properties of ultrasonic sensors

| Material | \(f_m\) /kHz | \(Z_m\) /\Omega | \(f_n\) /kHz | \(Z_n\) /\Omega | \(C_p\) /pF | \(\tan \delta\) | \(Q_m\) |
|----------|--------------|----------------|--------------|----------------|------------|---------------|--------|
| LFP0202  | 45.18        | 309            | 46.25        | 2.17           | 4.75       | 0.030         | 52.3   |
| PZT-5A   | 43.85        | 143            | 46.00        | 4.03           | 5.47       | 0.014         | 50.7   |
| PZT-4    | 44.45        | 151            | 45.98        | 5.21           | 4.31       | 0.002         | 84.2   |

3. Results and discussion

3.1 Piezoelectric properties of ultrasonic sensors

Table 3 shows the piezoelectric properties of the ultrasonic sensor prepared using each piezoelectric element. In the table, \(f_m\) and \(f_n\) are the minimum and maximum impedance frequencies, respectively, \(Z_m\) and \(Z_n\) are the impedance at \(f_m\) and \(f_n\), respectively, and \(C_p\) is the capacitance. The \(C_p\) and \(\tan \delta\) values are based on the material properties. All of the sensors have an \(f_m\) of approximately 45 kHz and a \(Q_m\) value within the range 50–80. Because characteristics such as \(f_m\) and \(Q_m\) are affected by the structure and material of the vibrating part, the characteristics of such a structure mainly approach the characteristics of the metal plate. The difference in \(Z_m\) and \(Z_n\) can be characterized by the difference in mechanical loss and electromechanical conversion efficiency, which PZT-5A and LFP0202 with equivalent \(Q_m\) are based on \(k_p\) and PZT-4 and LFP0202 with similar \(k_p\) are based on \(Q_m\).

3.2 Transmission/reception characteristics

Figure 2 shows the sound-pressure frequency characteristics as transmission characteristics, and Fig. 3 shows...
the sensitivity frequency characteristics as reception characteristics of the ultrasonic sensors fabricated using LFP0202, PZT-5A, and PZT-4. Table 4 shows the characteristic values from Figs. 2 and 3. The maximum SPL at 250 mW is approximately the same for the LFP0202 and PZT-5A devices and higher for the PZT-4 device. In terms of the half-width (Table 4), the PZT-5A device exhibits the widest bandwidth and the bandwidth tends to narrow in the order of LFP0202 and PZT-4.

If the sound-pressure characteristics are described only by the static characteristics $Q_m$ and $d_{31}$, the maximum SPL of the LFP0202 device is expected to be lower than that of the PZT-5A device, which has an equivalent $Q_m$ and a larger $d_{31}$. However, the LFP0202 device exhibited the same maximum SPL as that of the PZT-5A device. The bandwidth of PZT-5A device is wider than the especially increased vibration damping during driving; that is, the $Q$-factor in terms of the vibration during driving seems to be smaller for LFP0202. Therefore, the SPL of the ultrasonic sensor fabricated from PZT-5A was the same as that of the sensor fabricated from LFP0202 with a small $d_{31}$. The PZT-4 exhibits the highest SPL and narrow bandwidth because of its $Q_m$.

We here discuss the difference in frequency characteristics between LFP0202 and PZT-5A. Fig. 2 shows that the maximum sound-pressure frequency, which corresponds to the resonance frequency, shifts to the lower-frequency side as the applied power increases, especially in the case of PZT-5A. For reference, the rate of decrease of the maximum sound-pressure frequency with 12.5 mW of driving power is shown in Table 3; the shift magnitude differs by piezoelectric material. This tendency of the change is not observed only by the temperature change of the sensor. Such a decrease in the resonance frequency in the high vibration velocity region is generally explained as a softening of the vibrator by an elastic nonlinear phenomenon.18,19 The resonant frequency is empirically described by the Young’s modulus in addition to the structure, density, and Poisson’s ratio. Therefore, the difference in frequency characteristic behavior between PZT-based ceramics and LFP0202 can be attributed to differences in the vibration velocity dependence of material hardness. However, for these rigorous discussions it is necessary to compare in the higher vibration velocity range to see distinct data differences, which are not suitable for durability in this device.

According to Fig. 3, PZT-4 exhibits the highest sensitivity, followed by PZT-5A and LFP0202. The half-widths of LFP0202 and PZT-5A are approximately the same, and that of PZT-4 is the narrowest. Each maximum sensitivity frequency corresponds to the antiresonance frequency. This result can be sufficiently explained by the static-state $g_{31}$ in Table 1 and $Q_m$ values reported in Table 3. Moreover, we identified that dynamic characteristics such as sound pressure are affected by the mechanical character-

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**Table 4. Transmission/reception characteristics of ultrasonic sensors using LFP0202, PZT-5A and PZT-4**

| Material | Maximum SPL /dB | Half width /kHz | Frequency shift */% | Maximum sensitivity /dB | Half width /kHz |
|----------|----------------|----------------|-------------------|------------------------|----------------|
| LFP0202  | 120.8          | 1.55           | -0.44             | -65.6                  | 1.20           |
| PZT-5A   | 120.0          | 1.71           | -0.91             | -61.9                  | 1.10           |
| PZT-4    | 123.9          | 1.00           | -0.90             | -61.0                  | 0.95           |

1During 250 mW driving.

2Compared with 12.5 mW driving.
istics from transmission/reception. Furthermore, the difference between the maximum sensitivity of the LFP0202 and PZT-5A devices is only $-4 \text{ dB}$, suggesting that the LFP0202 sensor can be used in practical applications.

### 3.3 Temperature increase during driving

Figure 4 shows the results of measurements of the surface temperature of the ultrasonic sensor metal plate under an applied voltage. The surface temperature increases with increasing driving time, reaching a constant temperature within $\approx 180 \text{ s}$. At 250 mW, the LFP0202 and PZT-5A increase to similar temperatures, both of which are greater than that of PZT-4. The temperature increase during driving is attributed to $Q_m$ and $\tan \delta$, which represent the mechanical loss and the electrical loss, respectively. However, at 125 mW, LFP0202 exhibits a slow temperature increase and reaches a lower final temperature, although the relationship between PZT-5A and PZT-4 does not change. Table 4 shows no significant difference in the relationship between the sound pressure (stress) and the applied power between the LFP0202 and PZT-based ceramics. Thus, the difference in heat generation might be attributable to differences in material properties. We speculate that the difference in temperature increases at low loads is due to the slightly greater thermal conductivity of LFP0202 compared with that of the PZT-based ceramics.

### 3.4 Responsiveness of vibration velocity

Figure 5 shows the vibration velocity when a burst wave was applied. The applied voltage was set so that the vibration velocity during continuous driving was $1 \text{ m s}^{-1}$ (peak-to-peak). The vibration velocity was normalized by the maximum value of each sensor. The input condition of burst wave was set to 100 cycles, which corresponds to the time to reach the maximum vibration velocity during continuous driving. The results in Fig. 5 show that LFP0202 and PZT-5A respond with the same rising time and falling time, which are shorter than those of PZT-4. This trend is generally explained on the basis of $Q_m$. LFP0202 has a higher Young’s modulus and a lower density than the PZT-based ceramics (Table 1). Moreover, the results in Fig. 2 indicate the possibility of a difference in the behavior between PZT-5A and LFP0202 at high vibration velocity, even if $Q_m$ of static characteristics is nearly the same; thus, we did not expect to be able to explain the response trend on the sole basis of $Q_m$. However, because of vibration damping of the elastic body, the hardness (high $Y'$) and the lightness (low $\rho$) cancel each other; as a result, the LFP0202 exhibits a responsiveness equivalent to that of PZT-5A.

### 4. Conclusion

We fabricated an ultrasonic sensor using the KNN–NTK-based Pb-free piezoelectric ceramic LFP0202 and compared the sensor characteristics with those of sensors fabricated using PZT piezoelectric ceramics. The ultrasonic sensor fabricated using LFP0202 exhibited characteristics equivalent to those of the PZT-5A-based sensor in terms of sound pressure, sensitivity, temperature change during driving, and responsiveness of vibration velocity. PZT-5A is used for general ultrasonic sensors and has a $Q_m$ equivalent to that of LFP0202. Most sensor characteristics tend to be explained by static properties such as coupling coefficients ($d_{31}$, $g_{31}$) and losses ($Q_m$, $\tan \delta$). However, the resonance frequency shift with the broadening of sound-pressure characteristics in LFP0202 tended to be smaller than that of the PZT-based ceramics. This result explains why the LFP0202 sensor exhibit sound-pressure characteristics almost equivalent to those of the PZT-5A-based sensor, which has a larger $d_{31}$; this similarity suggests that the sound pressure in the nonlinear vibration region is dependent on material hardness.

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![Fig. 4. Temperature increase of ultrasonic sensors during driving. Solid line: 125 mW; dotted line: 250 mW.](image1)

![Fig. 5. Vibration velocity of ultrasonic sensors. Driving condition: 1 m$_{pp}$ s$^{-1}$, burst wave (100 cycles).](image2)
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