Effects of $K_4CuNb_8O_{23}$ and GeO$_2$ addition on microstructure and piezoelectric properties in KNbO$_3$ ceramics

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A combined additives study of lead-free KNbO$_3$ (KN) ceramics in the solid-state reaction was performed to improve the mechanical quality factor ($Q_m$) for high power piezoelectric applications. Improved $Q_m$ values of 1137 and 1241 were confirmed in KN specimens with addition of 1.0 mol\% K$_4$CuNb$_8$O$_{23}$ (KCN) and 1.0 mol\% KCN + 0.5 wt\% GeO$_2$, respectively. It is important for achievement of the higher $Q_m$ in KN systems to control the composition of the Cu segregated phase. In this study, the Cu segregated phase which was necessary to improve the $Q_m$ resulted from decomposition of the KCN compound during the sintering. Co-doping of KCN and GeO$_2$ to KN specimens has a remarkable effect in suppressing the deliquescence accompanying KCN addition and improvement of the $Q_m$ without significant deterioration of the electromechanical coupling factor $k$.$^3$. © 2019 The Japan Society of Applied Physics

1. Introduction

Piezoelectric devices at high power driving have widely been used for ultrasonic generators and piezoelectric transformers. Piezoelectric materials used for the high power piezoelectric application are preferably required not only to have high electromechanical coupling factor ($k$) and mechanical quality factor ($Q_m$) but also to have eminent mechanical fatigue resistance at high power driving.\(^1,2\) Currently, Pb(Zr$_x$Ti$_{1-x}$)$_3$O$_7$ (PZT) materials, which have excellent piezoelectric properties,\(^3\) are used in high power piezoelectric applications. In view of reducing damages to the environment from lead contained in PZT, however, the substitute demand for the lead-free piezoelectric materials has been actively conducted.\(^1,2\) Therefore, there are some limitations on the design of applications.

It has been studied to improve $Q_m$ for high power piezoelectric applications. Improved $Q_m$ values of 1137 and 1241 were confirmed in KN specimens with addition of 1.0 mol\% K$_4$CuNb$_8$O$_{23}$ (KCN) and 1.0 mol\% KCN + 0.5 wt\% GeO$_2$, respectively. It is important for achievement of the higher $Q_m$ in KN systems to control the composition of the Cu segregated phase. In this study, the Cu segregated phase which was necessary to improve the $Q_m$ resulted from decomposition of the KCN compound during the sintering. Co-doping of KCN and GeO$_2$ to KN specimens has a remarkable effect in suppressing the deliquescence accompanying KCN addition and improvement of the $Q_m$ without significant deterioration of the electromechanical coupling factor $k$.$^3$. © 2019 The Japan Society of Applied Physics

2. Experimental procedure

All of the KN ceramics under investigation were fabricated in a solid-state reaction. Powders of KHCO$_3$ (Takasugi Pharmaceutical Co., Ltd., Japan 99.9\%) and Nb$_2$O$_5$ (Mitsui Mining & Smelting Co., Ltd., Japan 99.9\%) were used as starting materials. These starting materials were weighed at a compositional ratio ($A/B$) of 0.994. It was confirmed before this research that sintered KN ceramics with this compositional ratio did not exhibit deliquescence. The compositional ratio of mixtures was analyzed with beaded KN powders by quantitative analysis of X-ray fluorescence using ZSX-100e (Rigaku Co., Ltd., Japan). Weighed powders were ball-milled in a 700 ml jar for 16 h and then were dried in an oven at 120 °C. Dried powders were separated into two parts. One
part was pressed into pellets of 60 mm diameter at 67 MPa and the other part was left unpressed to investigate physical properties of the calcined powders using the Brunauer–Emmett–Teller specific surface area (S) analyzed by Macsorb (MOUNTECH Co., Ltd., Japan) and diffractograms of X-ray diffraction (XRD) collected by SmartLab (Rigaku Co., Ltd., Japan) using a Cu Kα radiation. The unpressed powders exhibited tapped densities of 1.4 g cm⁻³.

These mixtures were firstly calcined at 600 °C for 4 h and then the top temperature of calcination was varied in the range of 750 °C–1030 °C for 4 h, as given in Fig. 1. The calcined grain size (d) was calculated from Eq. (1):

\[ d (\mu m) = 6/(S \times \rho), \]  

where \( \rho \) is the theoretical density of KN. The pH measurement was performed on solutions prepared by mixing 40 ml of deionized water with 0.2 g of calcined powders using F-24 (HORIBA, Ltd., Japan) calibrated with three standard buffer solution (4.01, 6.86 and 9.18 of pH, respectively). Refined lattice parameters of calcined powders were obtained using analysis software PDXL (Rigaku Co., Ltd., Japan) with a Crystallographic Information File (ICSD-190922).

Calcined powders of KCN were also synthesized using a solid-state reaction. Powders of KHCO₃, Nb₂O₅ and CuO (CIK Nanotek Co., Ltd., Japan 99%) were used as starting materials. Weighed powders were ball-milled in a 700 ml jar for 16 h and oven-dried at 80 °C. Dried powders were pressed into pellets of 60 mm diameter at 67 MPa and then calcined at 800 °C for 5 h. The calcined KCN powders were ball-milled and dried again under the same condition of the mixing process.

Then, 1.0 mol% KCN (KN + KCN), 1.0 mol% KCN and 0.5 wt% GeO₂ (Furuuchi Chemical Co., Ltd., Japan 99.999%) (KN + KCN + GeO₂) and 1.0 mol% CuO (KN + CuO) were added to KN pellets calcined at 1000 °C. These mixtures were ball-milled for 16 h and then oven-dried at 120 °C. Dried powders were mixed thoroughly with an acryl binder solution and then were pressed with 247 MPa into square pellets with a side length of 16 mm. All specimens were sintered for 2 h at 1046 °C after a de-binder process.

Densities of sintered specimens were measured by applying the Archimedes method. To examine the microstructure, the cross section areas of sintered specimens were polished with ethanol and then were observed by scanning electron microscopy (SEM) using S-4700 (Hitachi High-Tech. Corp., Japan). The post-deliquescence sample for SEM observation was obtained by repeating the procedure of dropping one drop of water on the polished surface and oven-drying at 60 °C from two to three times. Element mapping and a semi-quantitative point analysis in the cross section of the specimens were performed by electron probe micro analyzer (EPMA) using JXA-8500 (JEOL Ltd., Japan). The coefficient of variation (CV) of Cu concentration was calculated from Eq. (2):

\[ CV(\%) = 100 \times \sigma/\bar{x}, \]  

where \( \sigma \) and \( \bar{x} \) are the standard deviation and the mean value of Cu concentration, respectively. Inductively coupled plasma (ICP) analysis was carried out using ICP-8100 (SHIMADZU Corp., Japan). The irradiated supernatant for ICP analysis with ultrasonic waves for 30 min were made of 1.2 g of sintered specimens in 2 wt% ammonium oxalate.

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**Fig. 1.** Physical property values of bulk (empty maker and dashed line) and powder (filled maker and full line) calcined as a function of top temperature of calcination: (a) grain size (square symbols) and pH (circle symbols), (b) refined lattice parameter a, (c) refined lattice parameter b and (d) refined lattice parameter c.
In order to characterize dielectric and piezoelectric properties, silver paste was coated on both sides of 1 mm thick polished specimens and then these specimens were fired at 800 °C for 10 min to form the electrodes. The fired specimens were cut into appropriate shapes of (31) vibration mode. The temperature dependences of dielectric properties in the temperature range from 50 °C to 450 °C at 1 kHz were evaluated from an LCR meter using ZM2371 (NF Corp., Japan). The polarization versus electric field (P–E) hysteresis loops at 150 °C were measured with FCE-1 model (TOYO Corp., Japan). The specimens for the piezoelectric measurement were poled under a DC electric field of 3 kV mm$^{-1}$ at 150 °C in silicon oil for 15 min. All of the piezoelectric measurements were performed at least 24 h after polarization. $Q_m$ and $k_{31}$ were calculated on the basis of the EMAS-6100 standard from the resonance and anti-resonance frequencies provided by an impedance analyzer using HP4194A (Agilent Technologies, USA).

3. Results and discussion

3.1. Physical and piezoelectric properties

Grain size and pH value with KN calcined powders and bulks at various top temperatures from 750 °C to 1030 °C is shown in Fig. 1(a). Refined lattice parameter $a$, $b$ and $c$ are described at Figs. 1(b)–1(d), respectively. The diffraction peaks related to unreacted starting powders in all of the XRD profiles collected in this study were not confirmed. Increase or decrease of calcined grain size, pH value and lattice parameters up to 930 °C showed a same trend regardless of the filling condition of mixtures at the calcination. As the calcination temperature increased, the pH value decreased to the 7.5 and the grain size increased. The transition curve of the lattice parameters as a function of calcination temperature was consistent with the decrease curve of pH value which could extract the amount of unreacted potassium. Hence, the transition curve of lattice parameters indicates the diffusion behavior of the unreacted potassium into the KN grains. Namely, it is concluded that unreacted potassium has been present between grains up to 930 °C and played a role of forming a part of neck or liquid phases leading to the grain growth while diffusing into the KN grains.

There was no unreacted potassium in the samples calcined at a temperature higher than 930 °C. Therefore, the suppression of the grain growth above 930 °C up to 1030 °C via bulk calcination is understandable at the same manner as mentioned above. On the other hand, a rapid grain growth was only observed in the specimens with the powder calcination process, although there were no significant differences in lattice parameters of the calcined grains obtained via powder and bulk calcination. In order to understand the thermal behavior of grain growth via powder calcination, further investigation will be required. It was reported that it was difficult to increase the density of the sintered KN specimens which was calcined as powder. Therefore, the optimized calcination conditions$^{18,19,24,30}$ in which the top temperature of bulk calcination was fixed to 1000 °C were adopted to the calcination process in this additives study.

Refined lattice parameters $a$, $b$ and $c$ of the sintered KN specimens at 1046 °C are shown in Figs. 2(a)–2(c), respectively. There were no significant differences in all of the lattice parameters. Thus, the presence or absence of GeO$_2$ additives does not show a significant influence on lattice parameters. That is to say, it can be followed that the most part of the post-added GeO$_2$ was not inside the KN grains by solid-solution but was distributed at the grain boundaries.

Piezoelectric and physical properties from this research and previous reports are summarized in Table I. All specimens in this study showed a $T_c$ at 418 °C which was decreased by 6 °C in comparison with $T_c$ of un-doped KN.$^{24}$ Hence, it is concluded that the degrees of Cu solid-solution into the KN grains were almost the same and a small amount in all specimens in this research. All relative density values in this study were above 96%. A significant improvement of the $Q_m$ by doping of KCN to KN specimens was confirmed in this study. In addition, KCN doped KN specimens exhibited as high $Q_m$ as KCN doped KNN ceramics.$^{14,16}$ On the other hand, doping of CuO made the $Q_m$ of KN deteriorated greatly and the $Q_m$ values ($Q_m = 49$) and the sintering temperature ($T_s = 1046 °C$) in this study were different from the previous report ($Q_m = 611$, $T_s = 960 °C$).$^{19}$ Kim et al. also reported that the relative densities decrease at the sintering temperature over 980 °C. Hence, it might indicate that the powder calcination process was adopted to the previous study and has more effect for the $Q_m$ improvement in case of 1.0 mol% CuO doping.$^{19}$ In this research, it was not confirmed that the relative density made a
big impact on piezoelectric properties as in a previous report. Moreover, there were not significant differences in the grain information such as the lattice parameters and $T_c$ in all Cu additives doped KN specimens. However, piezoelectric properties were significantly different for each dopant. The cause of these differences will be discussed in Sect. 3.2.

A deliquescence was observed in this study with both KN $+$ KCN and KN $+$ KCN $+$ GeO$_2$ specimens which have promising piezoelectric properties, although the compositional ratio was lower than stoichiometric ratio as the previous report in KNN systems. The sintered body of KN $+$ KCN specimen was changed to powders when 72 h have been passed after the sintering. On the other hand, it should be stressed that the KN $+$ KCN $+$ GeO$_2$ specimen were not collapsed but turned the color of sintered body to white at the edges of sintered body after the specimen had been stored in atmosphere for a year. The delaying effect for the deliquescence with KN specimens by the post-addition of GeO$_2$ was observed without the deterioration of the $Q_m$ and the $k_{31}$. The cause and behavior, including the whitening, of the deliquescence will be discussed in Sect. 3.3. In contrast to the KCN doping, both of the enhancement of the $Q_m$ and the deliquescence were not observed in KN $+$ CuO specimen.

In short, the physical and piezoelectric properties of KN $+$ CuO showed the opposite behavior to KN $+$ KCN and KN $+$ KCN $+$ GeO$_2$.

### 3.2. Necessary condition of high $Q_m$ on microstructure

In order to explore the reason for the improvement effect of $Q_m$ in doping of KCN to the KN specimens in this study, further measurement and analysis were performed. Figure 3 shows the $P$-$E$ hysteresis loops of KN $+$ KCN $+$ GeO$_2$ equipped with high $Q_m$ and KN $+$ CuO equipped with low $Q_m$ at 150 °C which were the same as the temperature of the silicon oil at the poling condition. The rounded $P$-$E$ loop due to current leaks was observed in KN $+$ CuO specimen, however, both of the specimens demonstrated a ferroelectricity at 150 °C and the same coercive electrical field ($E_c$) of 1.7 kV mm$^{-1}$. This indicates the poling temperature and a DC electric field of 3 kV mm$^{-1}$ to perform the poling are not the main issue of deteriorated piezoelectric characteristics in KN $+$ CuO specimen. Hence, among the same material systems, the relationship with the $Q_m$ and the $E_c$ which had been reported before could not be adopted in this study.

The SEM images and the Cu element mapping of the sintered specimens in the cross section collected by EPMA analysis is depicted in Fig. 4. Segregations of the Cu element were verified in all of the fabricated specimens in this study. The degrees of Cu distribution were estimated using CV

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### Table I. Physical and piezoelectric properties of KN and KNN systems in this study and previous reports.

| Composition                        | Relative density (%) | $T_c$(°C) | $Q_m$ | $k_{31}$ | $k_{r}$ | $\theta_{max}$(°) | References     |
|------------------------------------|----------------------|-----------|-------|----------|--------|-------------------|----------------|
| KNbO$_3$ $+$ KCN 1.0 mol%          | 96.6                 | 418       | 1137  | 0.162    | —      | 81.6              | This study     |
| KNbO$_3$ $+$ KCN 1.0 mol% $+$ GeO$_2$ 0.5 wt% | 96.5                 | 418       | 1241  | 0.159    | —      | 82.1              | This study     |
| KNbO$_3$ $+$ CuO 1.0 mol%          | 96.5                 | 418       | 49    | 0.143    | —      | —                 | —              |
| KNbO$_3$ $+$ CuO 1.0 mol%          | 96                   | —         | 611   | —        | 0.29   | 86.9              | 31             |
| KNbO$_3$ $+$ CuO 1.0 mol%          | >96                  | 424       | 325   | 0.492    | 0.28   | 87.4              | 22             |
| KNbO$_3$ $+$ MnCO$_3$ 0.2 wt%      | 92.7                 | 282       | —     | 0.507    | —      | 87.2              | 30             |
| KNbO$_3$ $+$ MnCO$_3$ 0.8 wt%      | 96.1                 | 482       | —     | 0.43     | —      | 87.3              | 30             |
| $K_0.5Na_0.5NbO_3$ $+$ CuO 1.0 mol%| 97.5                 | 410       | 1210  | —        | 0.39   | 86–88             | 16             |
| $K_0.5Na_0.5NbO_3$ $+$ GeO$_2$ 0.5 mol% | 97.5                 | 410       | 1210  | —        | 0.39   | 86–88             | 16             |
| 0.94($K_0.5Na_0.5$)$_2$NbO$_3$ $+$ KCN 0.6 mol% | 97       | 440      | 983   | —        | 0.359  | —                 | 14             |
| 0.94($K_0.5Na_0.5$)$_2$NbO$_3$ $+$ KCN 1.0 mol% $>$96.7 | 431   | 792   | —      | 0.339   | —      | —                 | 14             |

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Fig. 3. (Color online) $P$-$E$ hysteresis loops of KN $+$ KCN $+$ GeO$_2$ and KN $+$ CuO.

Fig. 4. (Color online) SEM images and Cu element mapping of sintered specimens in cross section by EPMA analysis.
Thus, the necessary condition of improving the degrees of Cu distribution to avoid in value of Cu concentration that was inversely proportional to the degrees of Cu distribution to avoid influences from the nature of local analysis. The semi-quantitative analysis results at the points in the inset of Fig. 4 are summarized in Table II. Semi-quantitative analysis at Cu-segregated area was performed for 2 points at each specimen and the mean values of analysis results are shown in Table II. The content ratio of each element in Table II might comprise the analytical errors based on semi-quantitative analysis. There was no particular relationship between CV value of Cu and the Qm. There also were no significant differences in the compositional information of the KN grains themselves. Nevertheless, the compositional information of Cu segregation area in KN + CuO differed from the one in KN + KCN and KN + KCN + GeO2 Cu segregations. In particular, the oxygen occupancy (g_o) at the Cu segregation of KN + CuO differed from not only the one at the KN grains but also the one of KN + KCN and KN + KCN + GeO2. According to the previous reports in KN systems, the necessary condition for improving Qm was to substitute the Cu element at the B site of the perovskite structure as an acceptor to provide the pinning effect of defect dipoles.13) This indicates that most of the added CuO was localized in the segregated part had been the similar crystal structure as the one of the KN grains for substituted Cu element to act as an acceptor in the solid-solution.

Subsequently, the process of forming the Cu segregated part in the sintered specimens will be discussed. The composition at the Cu segregated area in KN + KCN and KN + KCN + GeO2 differ from a KCN itself. It can be speculated that the Cu segregated part were provided by the melting of the KCN around 1050 °C.31) On the other hand, in view of g_o, the compositional information at the Cu segregated area in KN + CuO was similar to the CuO itself. This indicates that most of the added CuO is localized in the sintered body while maintaining the crystal structure of CuO itself and some of it dissolve into KN grains. However, it is shown that the solid-solution of Cu element into the KN grains by the post-addition of Cu additives to the bulk calcined powders is not sufficient for increasing Qm as shown in Table II. Hence, it could be concluded that the Cu segregated parts which were necessary to improve the Qm were only provided by decomposing the KCN compound during sintering in this study.

3.3. Investigation of deliquescence
We performed the further investigation with respect to the deliquescence accompanying with the KCN post-addition to the KN specimens. The XRD profile patterns with calcined and sintered KN + KCN at 1046 °C before and after deliquescence are shown in Fig. 5. Although a homogeneous KN phase was obtained at the calcination process, K2Nb6O17 newly appeared in sintered specimens as a secondary phase. This observation can be understood if one considers the decomposing of the KCN compound during sintering. There were no significant differences in any crystallographic information before and after deliquescence. It could be concluded that the phenomenon of the deliquescence did not affect the grains themselves but the grain boundaries. The microstructure of the sintered KN + KCN before and after deliquescence at the same field in the cross section was investigated using SEM as shown in Figs. 6(a)–6(c). The elevated grains in the specimen after deliquescence were locally observed. From these results, it is concluded that the deliquescence phenomenon in KN systems is as the acting only on the grain boundaries and left to decomposition of the sintered body. There are three kinds of the state of the grain boundaries in Fig. 6(c). It can be interpreted that the swollen parts indicated by the blue arrow were the results of the aqueous solution progression in part of the grain boundary indicated by the black arrow. Hence, it can be speculated that the cavity parts at the grain boundary indicated by the red arrow occurred due to the generated local distortion from weakening of the bonding force at the grain boundary where the aqueous solution was formed. As a result, the elevated grains locally appeared on the polished surface after deliquescence. In order to reveal the compositional information of the swollen parts, the supernatant which was made of

| Composition | KN + KCN | KN + KCN + GeO2 | KN + CuO |
|-------------|----------|-----------------|---------|
| CV of Cu (%) | 62.7     | 139.2           | 123.2   |
| Element     |          |                 |         |
| K           | 20.9     | 21.6            | 19.6    |
| Nb          | 16.1     | 16.2            | 16.8    |
| Cu          | 6.4      | 2.4             | 15.6    |
| Ge          |          | 3.3             |         |
| O           | 56.6     | 56.5            | 47.7    |

Fig. 5. (Color online) XRD profile patterns with calcined and sintered KN + KCN specimens at before and after deliquescence.
After deliquescence.

Fig. 7 and Table III, respectively. of dissolved elements in aqueous solution containing ammonium oxalate are shown in Fig. 7 and Table III, respectively. The elution ratio of elemental composition and the total amount sintered specimens was analyzed by ICP analysis. The 2 wt% ammonium oxalate aqueous solution and 1.2 g of KN specimens was analyzed by ICP analysis. The elution ratio of elemental composition detected by ICP analysis.

Fig. 6. (Color online) SEM images with sintered KN + KCN specimens in cross section (a) before and (b) after deliquescence. (c) Enlarged SEM compositional image with sintered KN + KCN specimens in cross section after deliquescence.

The ratio of eluted potassium to potassium of added KCN decreased from 54.8% in KN + KCN to 0.6% in KN + KCN + GeO2. Therefore, it was found that the addition of the GeO2 to the KN specimens has a remarkable effect in suppressing the deliquescence without deterioration of the $Q_m$ and the $k_{31}$. It could be expected that most of the high water-solubility at the grain boundaries of KCN doped KN changed to water-insolubility such as K2GeO3 systems by post-added GeO2.

4. Conclusions

In order to improve the $Q_m$, a study of KCN, GeO2 and CuO post-addition to solid-state-reaction KN ceramics was performed. The behavior of grain growth at the phase of the calcination over 930 °C in KN mixtures differed between the bulk calcination and the powder calcination. All relative densities of the fabricated specimens using bulk calcination powders which did not contain unreacted potassium and did not show abnormal grain growth were above 96%. The highest $Q_m$ of 1241 reported for the KN systems up to now was found in KN specimens with 1.0 mol% KCN + 0.5 wt% GeO2 added.

The necessary condition of improving the $Q_m$ in the KN systems was that crystal structure of the Cu segregated part in the KN specimens is similar to one of the KN grains. The Cu segregated parts were only provided by decomposing the KCN compound during sintering in this study. Namely, via bulk calcination process in KN systems, the solid-solution of Cu element into the KN grains by the post-addition of Cu additives is not sufficient for increasing $Q_m$. Decomposing of the KCN compound during sintering as mentioned in Sect. 3.2. Thus, it could be concluded that one of the decomposed component from KCN was formed as the similar crystal structure as the KN ceramics and the other was formed as a high hydration activity component at the grain boundary.

| Table III. Total amount of dissolved elements and ratio of eluted potassium to potassium of added KCN amount detected by ICP analysis. |
| Composition | Total amount of dissolved element (mg) | Ratio of eluted K to K of added KCN amount (%) |
| KN + KCN | 6.220 | 54.8 |
| KN + KCN + GeO2 | 0.504 | 0.6 |
| KN + CuO | 0.020 | — |

Namely, the cause of the deliquescence accompanying with the addition of KCN was concluded that the grains and the grain boundaries in the specimens have different composition, and the remained components at the grain boundaries easily cause a hydration reaction with atmospheric moisture. Furthermore, the whitening at the surface of the sintered body is an indication of the deliquescence which can be ascribed mainly to the irregular reflection of the light at the cavity parts of the grain boundaries.

2 wt% ammonium oxalate aqueous solution and 1.2 g of sintered specimens was analyzed by ICP analysis. The elution ratio of elemental composition and the total amount of dissolved elements in aqueous solution containing ammonium oxalate are shown in Fig. 7 and Table III, respectively. The total amount of dissolved element has good correlation with the degrees of the deliquescence in the synthesized specimens. There was no detectable trace of the potassium from KN + CuO specimen which did not show the deliquescence. Therefore, in this ICP analysis, it is to be underlined that adjustment of the aqueous solution of ammonium oxalate does not affect the KN grains regardless of the presence or absence of deliquescence, and only the deliquescent component existing at the grain boundary was extracted. The aqueous solution of KN + KCN specimen contained all elements contained in the specimen. This elemental ratio including the potassium higher than 90% was neither KN nor KCN. This observation could be also interpreted that the post-added KCN compound decomposed during the sintering as mentioned in Sect. 3.2. Thus, it could be concluded that one of the decomposed component from KCN was formed as the similar crystal structure as the KN ceramics and the other was formed as a high hydration activity component at the grain boundary.

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It was found that the co-doping of KCN and GeO2 to the KN specimens has a remarkable effect in suppressing the deliquescence and improvement of the $Q_m$ without significant deterioration of the $k_{31}$. However, the addition of GeO2 could not completely eliminate the emergence of deliquescence accompanying the KCN doping.

As a future task, an investigation of controlling the composition at the Cu segregation area without an addition...
of KCN to the KN specimens is strongly desired in order to simultaneously achieve a further improvement of the $Q_m$ and the complete elimination of the deliquescence. A piezoelectric characterization at high power driving should be performed with optimized KN ceramics for high power applications.

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