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Hardening and magnetic field orientation behavior of KNN-based lead-free piezoelectric ceramics

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1. Introduction

Among piezoelectric materials, the PZT system is widely used in actuators, sensors, and ultrasonic transducers. However, PZT contains a significant amount of harmful lead. This has prompted the pursuit of lead-free alternatives[1)]. A lot of studies have identified $(K,Na)NbO_3$ (KNN) as one of the most promising lead-free alternatives inspired by the groundbreaking publication by Saito et al. 20 years ago[2)]. A large number of patents were filed simultaneously. To avoid patent issues, many studies have explored complex compositions. The validity period of patents that were filed has begun to expire over time.

Therefore, we focused on simpler material compositions and the hardening of KNN by application such as ultrasonic motors, transformers and ultrasonic power devices. KNN fabricated by conventional solid phase reaction from K_2CO_3, Na_2CO_3, Nb_2O_5 easily shows deliquescent, low insulation resistance, and low reliability, because the deficiencies or surpluses of alkali metal ions from the stoichiometric composition cause these problems. In this study, we fabricated $K_{0.5}Na_{0.5}NbO_3$ (KNN) and investigated the hardening of KNN by adding Ag_2O and CuO, which Ag forms $(K,Na,Ag)NbO_3$ and K-Na-Ag-O in deficiencies and surpluses, respectively and Cu acts acceptor ions for hardening of KNN system[3-6)]. In the previous study, the piezoelectric properties of $(K,Na,Li)NbO_3$ were enhanced by fabricating crystal-oriented ceramics using the high magnetic field method.[7)] Furthermore, we investigated the crystal-oriented behavior KNN system under high magnetic field.

2. Experimental Procedure

KNN powder was synthesized by conventional solid-phase reaction using regent-grade raw materials of K_2CO_3, Na_2CO_3, and Nb_2O_5. After mixing KNN powder with Ag_2O, CuO, the powders were pressed into the disks. The ceramic disks were obtained to sinter at 1100 to 1150°C for 2h and had dimensions ϕ8 x 0.5mm. Electrodes were formed on the surface of the disks using the silver printing technique, and the disks were poled at 150°C for 15 min under an electric field of 4 kV/mm. The dielectric and piezoelectric properties were evaluated such as dielectric constant and loss, electromechanical coupling factor, mechanical quality factor, and piezoelectric constant. Then the crystal-oriented behavior was evaluated to mold the dispersed powder such as slurry under the high magnetic field of 5T by the superconducting magnet (JASTEC JMTD-5T52M).

3. Results and Discussion

The relative density of KNN ceramics increases to over 97-98% with the addition of Ag_2O and CuO as shown in Figure 1. Ag_2O and CuO acted KNN as sintering aids. The electromechanical coupling factor k_r and the mechanical quality factor Q_m increased with the additives in Fig.2. Especially, Q_m increased exceeding 1200 with 0.02 of Cu content. KNN has almost single phase with Ag_2O to compensate K and Na sites of KNN. And then, KNN was hardening by additive of CuO.

Figure 3 represents that KNN powder was oriented along the b and c- axes parallel to the magnetic field. Therefore, it is expected that the piezoelectric properties of KNN system were enhanced by crystal orientation applying high magnetic field.

References

1) A. Safari, Q. Zhou, Y. Zeng, and J. D. Leber, Jpn. J. Appl. Phys. **62**. SJ0801 (2023).
2) Y. Saito, H. Takao, T. Tani, T. Nonoyama, K. Takatori, T. Homma, T. Nagaya, and M. Nakamura, Nature **432**, 84 (2004).
3) https://oqmd.org/materials/composition/Na2Ag6O4.
4) https://oqmd.org/materials/composition/K2Ag2O2
5) R. Umamaheswari, M. Yogeswari, G. Kalpana,

SolidStateCommun. **155** 62 (2013).
6) M. Matsubara, T. Yamaguchi, K. Kikuta, and S. Hirano, Jpn. J. Appl. Phys. **44**, 6136 (2005).
7) Y. Ono, T. Harada, H. Shimizu, Y. Doshida, T. Honma, and S. Tanaka, J. Ceram. Soc.Jpn. 127, 887 (2019)

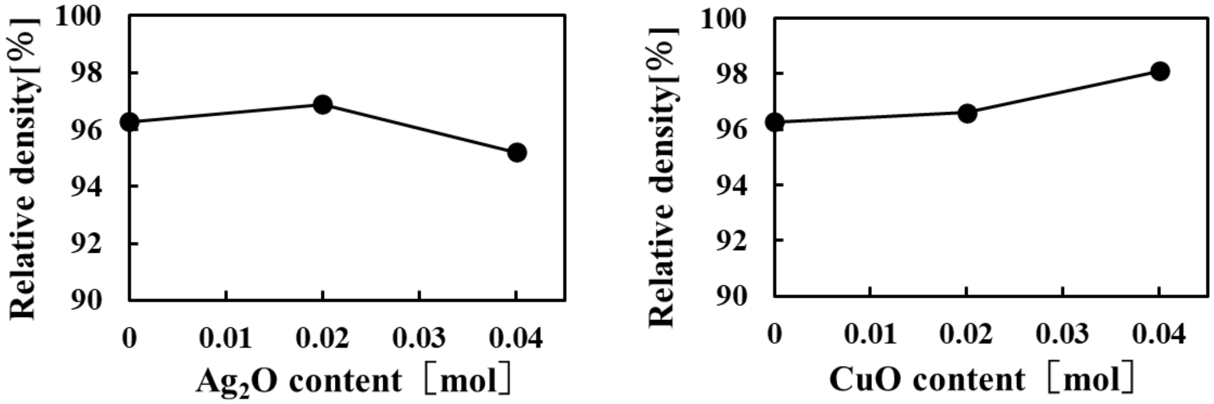

Fig.1 Relative density vs additive content characteristics.

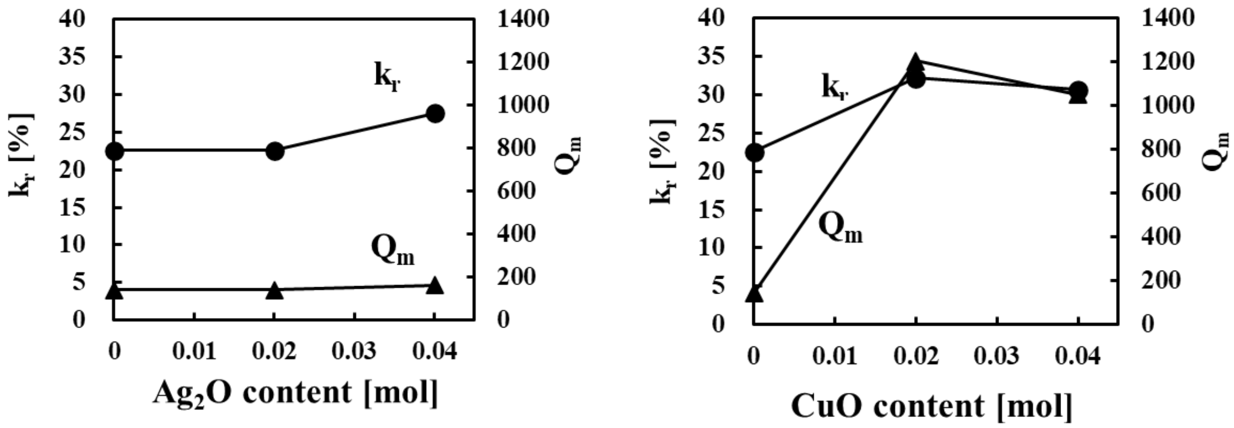

Fig.2. Piezoelectric properties vs additive content characteristics.

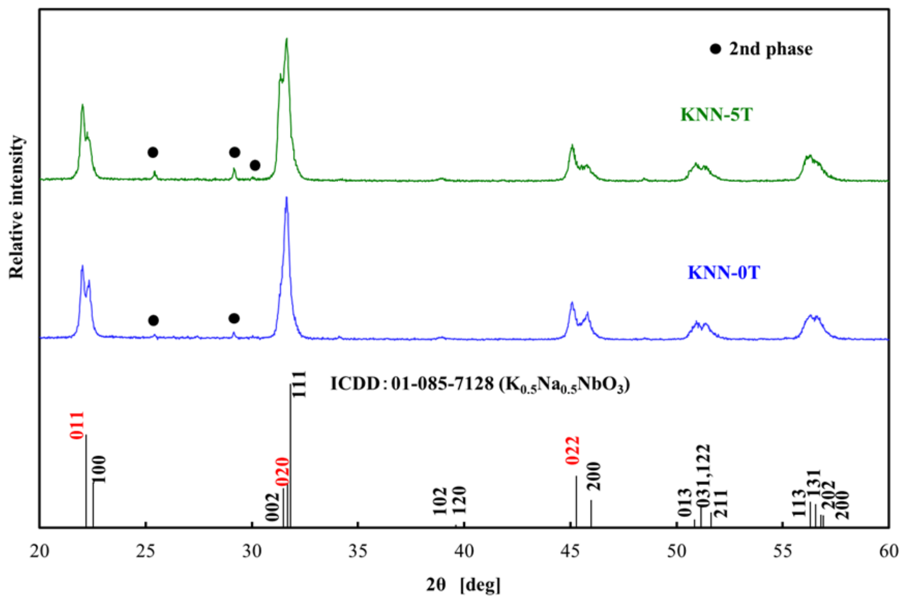

Fig.3. XRD profiles with or without standing magnetic field.