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Effects of LiF on the Structure and Electrical Properties of $(Na_{0.52}K_{0.435}Li_{0.045})Nb_{0.87}Sb_{0.08}Ta_{0.05}O_3$ Lead-Free Piezoelectric Ceramics Sintered at Low Temperatures

P. D. Gio*, N. D. Phong

Department of Physics, College of Sciences, Hue University, Hue, Vietnam
Email: *pdg_55@yahoo.com

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Abstract

The $(Na_{0.52}K_{0.435}Li_{0.045})Nb_{0.87}Sb_{0.08}Ta_{0.05}O_3$ (KNNLST) + x wt% LiF piezoelectric ceramics, where x = 0, 2, 4, and 6, have been fabricated successfully by the conventional solid-state reaction method. The effect of LiF on the sintering temperature, the structure and electrical properties of KNNLST ceramics was systematically studied. The LiF addition significantly reduced the sintering temperature of the ceramics from 1100˚C to 930˚C. Experimental results showed that with the doping of LiF, all the ceramic samples could be well sintered and exhibit a dense, pure perovskite structure. With increasing LiF content, the tetragonal-orthorhombic transition point (T_{O-T}) and the Curie temperature (T_c) of the ceramics shifted to the lower and higher temperatures, respectively. The specimens containing 4 wt% LiF sintered at 930˚C showed the good electrical properties: the density of 4.26 g/cm^3; the electromechanical coupling factor, k_p = 0.27 and k_t = 0.40; the dielectric constant, ε = 744; the piezoelectric constant (d_{31}) of 41 pC/N.

Keywords

Lead-Free, Crystal Structure, Piezoelectric Properties, Electromechanical Coupling Factor

1. Introduction

Although the piezoelectric ceramic materials on the basis of PZT have the perfect physical properties and have

*Corresponding author.

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many important applications in science and technology such as in sensors, actuators and the other electronic devices [1]-[6], they contain large amounts of toxic lead more than 60 wt%, their high volatilization and toxicity of PbO during firing process can contaminate the environment and damage human health. So, the replacement of lead by lead-free materials for piezoceramics is essential.

During recent years, intensive efforts have been made to develop lead-free piezoelectric ceramics such as $BaTiO_3$, $Na_{0.5}Bi_{0.5}TiO_3$, $(Bi_{0.5}Ka_{0.5})TiO_3$, $(K,Na)NbO_3$, etc. [7]-[12]. Among them, $(K, Na)NbO_3$ (KNN) based piezoelectric ceramics was the most interested because of its strong piezoelectric properties and high Curie temperature [7] [13]-[16]. However, the sintering temperatures of KNN based ceramics fabricated by conventional methods are quite high (>1000˚C), this leads to evaporation of alkaline elements during sintering process, resulting in the reduced properties of ceramics. Therefore, lowering sintering temperature of KNN based ceramics is very important.

Many researchers have successfully decreased the sintering temperature of lead-free ceramics by using the low-temperature melting additives such as Bi_2O_3, Li_2CO_3, CuO, LiF, etc. [17]-[22]. In some cases, these additives can reduce easily the sintering temperature, but also reduce the piezoelectric properties of ceramics due to the appearances of second phase, or variation of structure phase [17]. Currently, the research and manufacture of ceramics material sintered at a low temperature, which leads to improvement or does not reduce the piezoelectric properties of ceramics system, is very interesting.

In this work, we present some research results on the effect of LiF on the sinterability, structure and electrical properties of $(Na_{0.52}K_{0.435}Li_{0.045})Nb_{0.87}Sb_{0.08}Ta_{0.05}O_3$ ceramics. The LiF dopant significantly reduced the sintering temperature and enhanced the electrical properties of the ceramics at room temperature.

2. Experimental Procedure

The $(Na_{0.52}K_{0.435}Li_{0.045})Nb_{0.87}Sb_{0.08}Ta_{0.05}O_3$ (KNNLST) + x wt% LiF ceramics, where x = 0, 2, 4 and 6, were synthesized by a conventional mixed-oxide method. Potassium carbonate (K_2CO_3), sodium carbonate (Na_2CO_3), lithium carbonate (Li_2CO_3), and oxides Sb_2O_5, Nb_2O_5, Ta_2O_5 (purity ≥99%) were used as starting materials. Mixed powder KNNLST was milled for 20 h with the ZrO_2 balls in ethanol. Two calcinations at temperature 850˚C for 2 h were then performed to obtain the single phase formation. Thereafter lithium fluoride (LiF) was mixed with the calcined KNNLST powder, and powders milled for 16 h. The ground materials were pressed into disk 12 mm in diameter and 1.5 mm in thick under 100 MPa. To prevent the evaporation of alkaline elements, these pellets were covered by the powders with the same composition and then were sintered in a sealed alumina crucible at the temperature of 900˚C, 930˚C, 950˚C, and 1000˚C for 2 h.

The crystal structure of the sintered samples was examined by X-ray diffraction (XRD, D8 ADVANCE). The microstructure of the samples was examined by using a scanning electron microscope (SEM) (Hitachi S-4800). The densities of samples were measured by Archimedes method. The ceramic samples were polarized in silicone oil at 120˚C by applying dc field of 30 kV/cm for 20 min, then cooled down to room temperature under the same electric field. They were aged for 24 h prior to testing. The piezoelectric properties were determined by the resonance and antiresonance frequencies using an impedance analyzer (HP 4193A and RLC HIOKI 3532). Temperature dependence of dielectric constant was determined using RLC HIOKI 3532 with automatic programming; temperature of the samples was measured using Digital Multimeter 7562. The ferroelectric property was measured by Sawyer-Tower method.

3. Results and Discussion

3.1. Sintering Behavior

Figure 1 shows the densities of KNNLST ceramics with 4 wt% LiF content as a function of sintering temperature. With increasing of sintering temperature, the density increases and reaches the maximum value (4.26 g/cm^3) at 930˚C sintering temperature, then decreases. Meanwhile, the sintering temperature of undoped KNNLST ceramics was as high as 1100˚C, the density of 4.24 g/cm^3 (**Table 1**). Thus, the addition of LiF improved the sinterability, reduced the sintering temperature of 170˚C compared with pure samples and increasing density of the ceramic samples. This is probable due to the formation of liquid phase of LiF. As is well known, LiF has a low melting point of 845˚C, therefore it may form liquid phase during sintering process, which could probably promote the densification behavior of KNN-based piezoceramics at reduced temperatures [17].

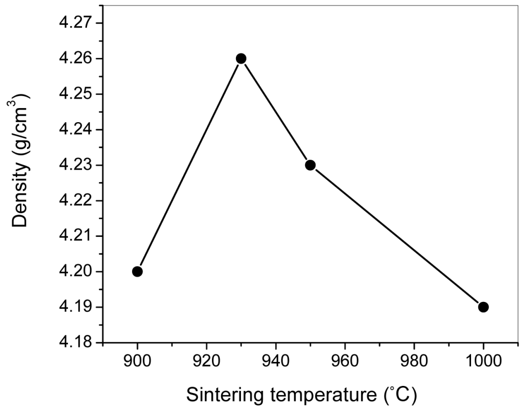

Figure 1. The density of KNNLST + 4 wt% LiF ceramics sintered at different temperatures: 900˚C, 930˚C, 950˚C, and 1000˚C.

Table 1. Some properties of undoped KNNLST (sintered at 1100˚C) and KNNLST + 4 wt% LiF (sintered at 930˚C) ceramics.

Compositions	Sintering temperature (˚C)	Density (g/cm^3)	k_p	k_t	ε
KNNLST	1100	4.24	0.25	0.32	855
KNNLST + 4 wt% LiF	930	4.26	0.27	0.40	744

Figure 2 shows the planar electromechanical coupling factor k_p, k_t and dielectric constant ε of KNNLST + 4 w% LiF ceramics as a function of sintering temperature. The k_p, k_t and ε show similar tendency, initially they increase with the sintering temperature, reach the maximum values at 930˚C sintering temperature, then decrease when sintering temperature exceeds 930˚C. As indicated in **Table 1**, for the KNNLST + 4 w% LiF ceramics sintered at 930˚C showed the good electrical properties, the electromechanical coupling factor k_p, k_t are 0.27, 0.40, respectively, which were relatively higher than that pure KNNLTS ceramics. These results may be related to the suppressed volatilization of alkali elements at the low temperature sintering and the increase of density as discussed above.

From the results above, the sintering temperature for KNNLST doped LiF ceramics was selected as 930˚C.

3.2. Structure, Microstructure and Electrical Properties

Figure 3(a) shows the XRD patterns at room temperature with the 2θ range from 20˚ to 70˚ of KNNLST + x wt% LiF ceramic samples sintered at 930˚C. It can be seen that all samples exhibit dominant phase structure of perovskite ABO_3 type, beside a small amount of second phase $LiSbO_3$ (PDF card # 024-0598) and $K_6Nb_{10.8}O_{30}$ (PDF card # 070-505) have been found in pure KNNLST and KNNLST doped 2 w% LiF samples as shown in **Figure 3(a)**. The appearance of second phase may be due to the composition inhomogeneity of ceramics [17]. Crystal structure of the samples is modified significantly by LiF additions as shown in **Figure 3(b)**.

Figure 3(b) shows the enlarged XRD patterns of the ceramics in the ranges of 2θ from 45˚ to 47˚. It is well known that KNN-based ceramics usually show orthorhombic symmetry at room temperature [14]. According to research results of the works [17] [22] [23], when the ceramics is of orthorhombic phase, the relative intensity of (002)/(020) peaks is approximately 2 and the (002) peak appears at a smaller Bragg angle, but the I(002)/I(020) decreases to 0.5 for a tetragonal phase. Therefore, from X-ray diffraction diagram in **Figure 3(b)** shows that likely the dominate phase in the pure KNNLST ceramics is orthorhombic, whereas the addition of LiF changes it to tetragonal symmetry. With x equals 4, the ceramics shows the coexistence of two phases, and then with x = 6, the tetragonal phase became dominant. This result suggests that may be Li^+ ions are substituted for A-site of perovskite structure ABO_3 which leads to the change of crystal structure from orthorhombic to tetragonal. So it could be said that the composition with x = 4 is near to the morphotropic phase boundary of KNNLST doped LiF ceramics [16].

Figure 4 shows the SEM images of the KNNLST + x wt% LiF ceramics sintered at 930˚C: (a) x = 0, (b) x = 2, (c) x = 4 and (d) x = 6. It can be seen that the microstructure of the pure KNNLST ceramics with square or

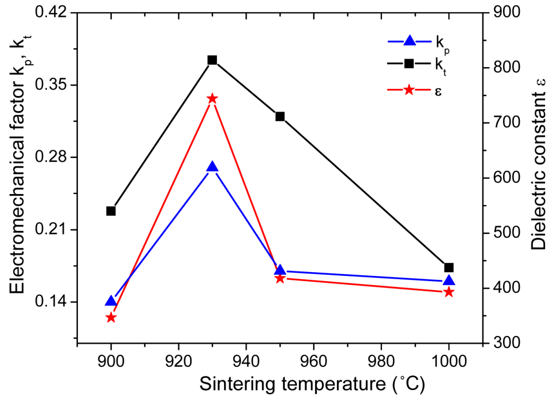

Figure 2. Electromechanical coupling factor k_p, k_t and dielectric constant ε of KNNLTS + 4 w% LiF ceramics as a function of sintering temperature.

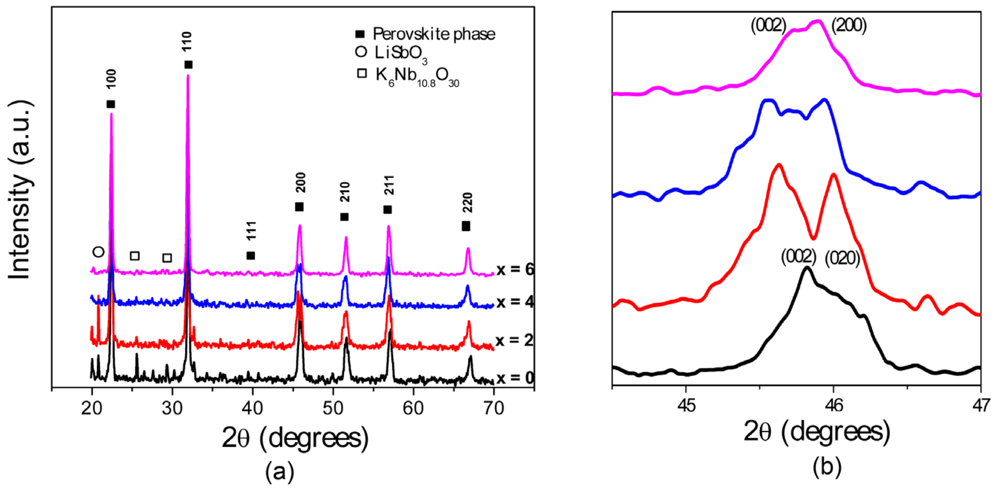

Figure 3. XRD patterns of the KNNLTS + x wt% LiF ceramics (a) and enlarged region for (002)/(020) peaks of the diffraction patterns (b).

rectangular shaped grains discrete distributions, porous, not sintered materials as shown in **Figure 4(a)**. However, the microstructure of samples becomes denser and average grain size increases from 0.4 to 2 μm (shown in **Table 2**) as the LiF content is increased (**Figures 4(b)-(d)**). A homogeneous microstructure developed for the sample with 4 wt% LiF added (**Figure 2(c)**). **Figure 4(d)** also shows that further increasing LiF content to 6 wt% gives rise to some large abnormal grains, porous. Such with the 4 wt% LiF added sample, the highly dense and homogeneous microstructure was obtained, which may expect improved properties of ceramics. The grain growth with LiF addition can be explained by liquid phase sintering.

Figure 5 shows temperature dependence of dielectric constant ε and dielectric loss tanδ measured at 1 kHz of KNNLST + x wt% LiF ceramics sintered at 930˚C. As seen in **Figure 5**, KNNLST doped LiF ceramics undergo two phase transition, that are the orthorhombic-tetragonal phase transition (T_{O-T}) and tetragonal-cubic phase transition (T_C) during the measured temperature range [16] [17] [22] [23]. **Figure 6** shows the orthorhombic-tetragonal phase transition temperature (T_{O-T}) and Curie temperature (T_C) of KNNLST ceramics sintered at 930˚C change as a function of LiF content. The pure KNNLST ceramics possess T_{O-T} and T_C temperature of 67˚C and 193˚C, respectively. With the addition of LiF, the T_{O-T} temperature of KNNLST ceramics decreases consistent with the analysis of XRD patterns that the phase structure of ceramics changes toward tetragonal symmetry when LiF content increases, while T_C temperature increases due to diffusion of Li^+ into A-site of KNN lattice, showing similar tendency to the works [17] [23]. **Figure 5** also shows that the dielectric loss little changes as the temperature rises from room temperature to 150˚C and increases rapidly in the vicinity Tc.

Some physical properties of KNNLST+ x wt% LiF ceramics sintered at 930˚C measured at room temperature

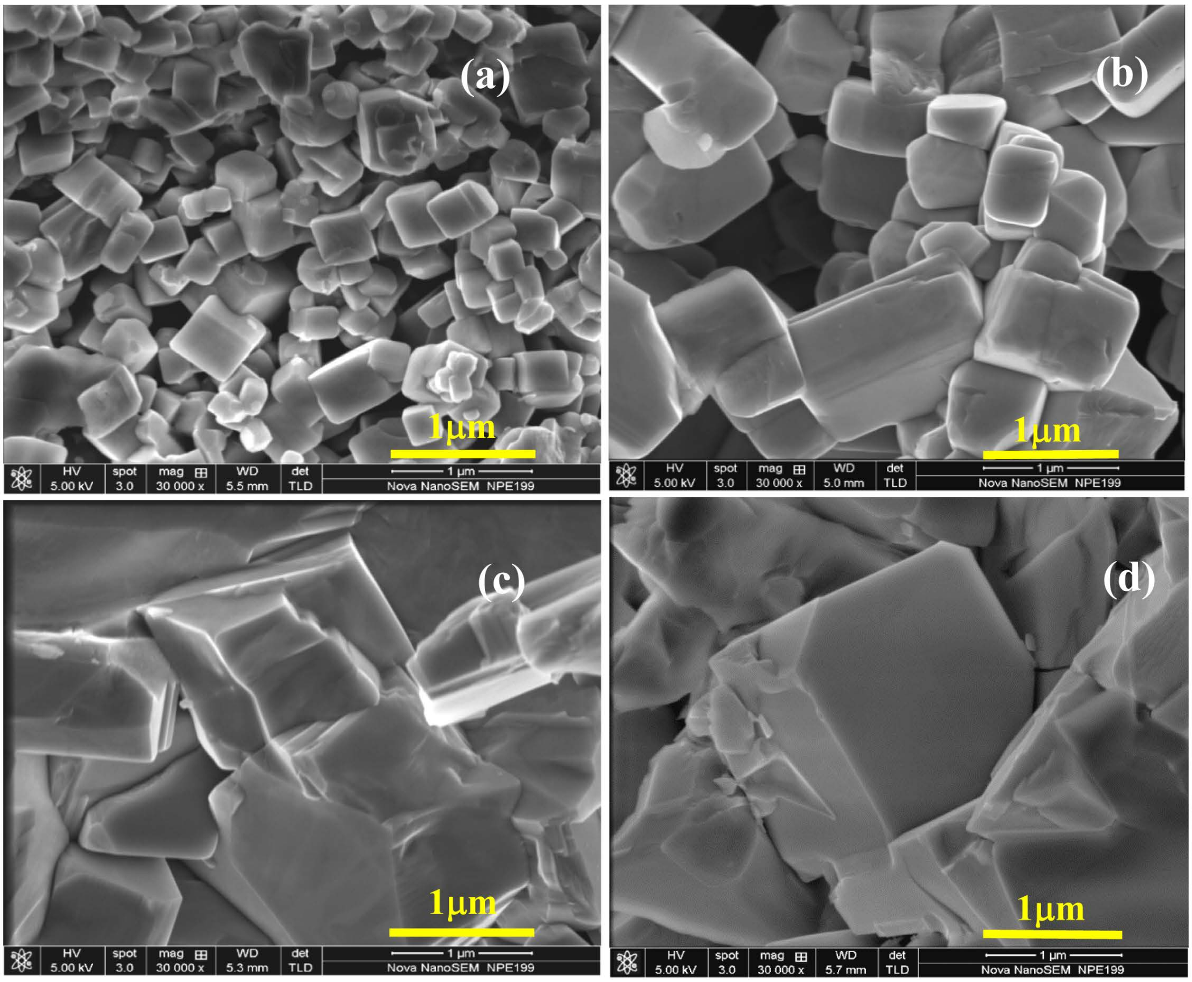

Figure 4. SEM images of KNNLST + x wt% LiF ceramics sintered at 930˚C: (a) x = 0; (b) x = 2; (c) x = 4; (d) x = 6.

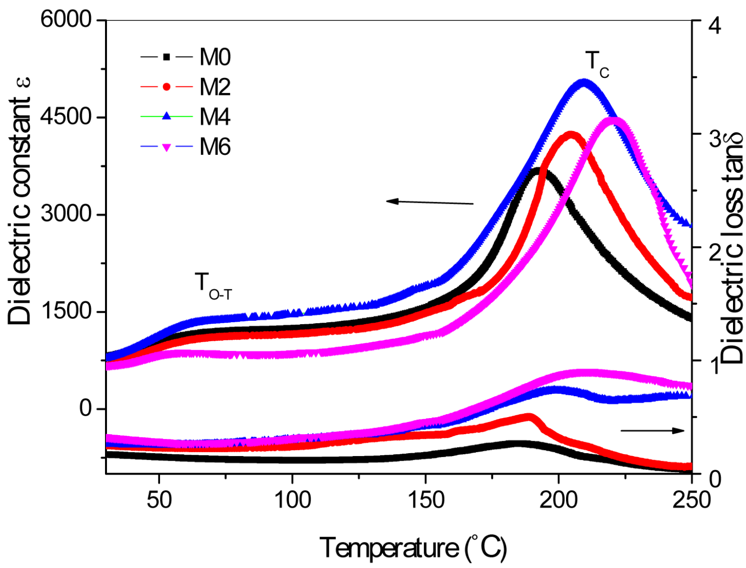

Figure 5. Temperature dependence of dielectric constant ε and dielectric loss tanδ of the KNNLST + x wt% LiF ceramics sintered at 930˚C.

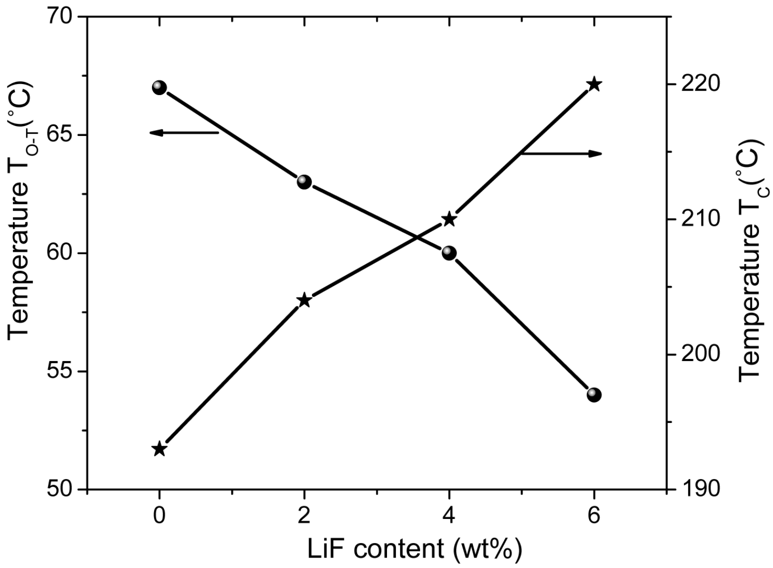

Figure 6. The orthorhombic-tetragonal phase transition temperature (T_{O-T}) and Curie temperature (T_C) of KNNLST ceramics as a function of LiF content.

Table 2. Physical properties of KNNLST + x wt% LiF ceramics sintered at 930˚C.

x content	Density (g/cm^3)	Average grain size (μm)	ε	k_p	k_t	$-d_{31}$ (pC/N)
0	4.03 ± 0.01	0.4 ± 0.15	470	0.12	0.29	15
2	4.15 ± 0.01	0.8 ± 0.1	577	0.16	0.36	23
4	4.26 ± 0.02	1.7 ± 0.3	744	0.27	0.40	41
6	4.16 ± 0.01	2.0 ± 0.2	650	0.17	0.28	20

also listed in **Table 2**. It is seen that with increasing of LiF content, the density, dielectric constant (ε), planar electromechanical coupling factor (k_p), (k_t), piezoelectric constant (d_{31}) are increased. The largest values for density of 4.26 g/cm^3, k_p of 0.27, k_t of 0.40, d_{31} of 41 pC/N, ε of 744 were obtained at 4 wt% LiF content, then decrease. These are probably related to characteristics of the density, the increasing grain size and morphotropic phase boundary effect [16].

Figure 7 shows the shape of ferroelectric hysteresis loops of the samples KNNLST+ x wt% LiF measured at room temperature. From the shape of these loops, the remanent polarization P_r and the coercive field E_C of ceramics were determined, as shown in **Figure 8**. With increasing of LiF content, a sharp increases in P_r was observed for samples until x = 4, reaches the highest value (11.96 μC/cm^2) at this content, and then decreases; while the coercive field E_C strong decreases from 8.06 to 5.38 kV/cm during the measured LiF content range. These results are in good agreement with the studied dielectric and piezoelectric properties of the ceramic samples.

4. Conclusion

The effect of LiF addition on the sintering behavior and physical properties of $(Na_{0.52}K_{0.435}Li_{0.045})Nb_{0.87}Sb_{0.08}Ta_{0.05}O_3$(KNNLST) + x wt% LiF (x = 0 ÷ 6) ceramics was investigated. The addition of LiF improved the sinterability of the ceramics and caused an increase in the density and grain size at low sintering temperature (930˚C). All samples had perovskite phase structure with a change from orthorhombic to tetragonal by LiF additions and a possible morphotropic phase boundary near composition x = 4. With increasing LiF content, the T_{O-T} temperature of the ceramics was shifted to lower temperature, while the Curie T_C temperature was shifted to higher temperature. At the LiF content of 4 wt%, physical properties of ceramics sintered

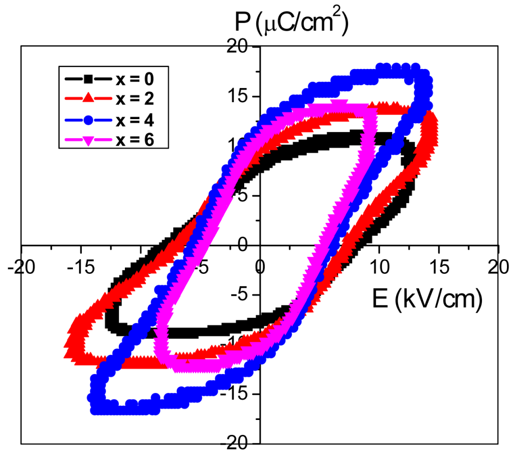

Figure 7. Hysteresis loops of KNNLTS + x wt% LiF ceramic samples measured at room temperature.

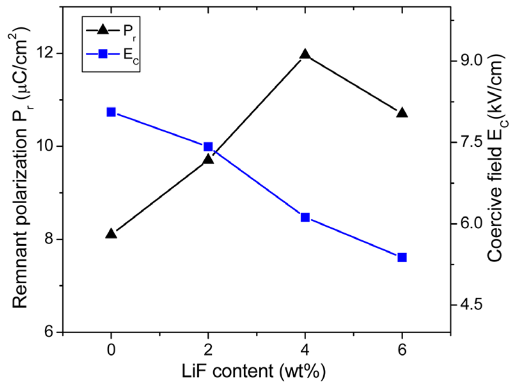

Figure 8. The remnant polarization (P_r) and coercive field (E_C) of KNNLST ceramics as a function of the LiF content.

at 930˚C are the best.

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