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Enhanced piezoelectric properties of (Bi,Na)TiO_3–(Bi,K)TiO_3 ceramics prepared by two-step sintering process

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Abstract

To enhance the piezoelectric properties and lower the sintering temperature of lead-free (Bi,Na) TiO_3-(Bi,K) TiO_3 ceramics, two-step sintering process was employed and investigated. As a representative lead-free piezoelectric materials, (Bi,Na) TiO_3-(Bi,K) TiO_3 has been selected for the two-step sintering process. The two-step sintering process can be the advisable fabrication process, especially for the case of volatile materials were contained. The two-step sintering process can also minimize the evaporation of volatile components. By lowering the sintering temperature and increasing the sintering time in the second step, grain size was enlarged and density was improved. By optimizing the holding time from the $0.80Bi_{0.5}Na_{0.5}TiO_3$-$0.20Bi_{0.5}K_{0.5}TiO_3$ ceramics, improved bulk density of 5.81 g/cm^3, piezoelectric charge coefficient of 170 pC/N and dielectric constant of 1471.29 were obtained. As a result, piezoelectric and structural properties can be enhanced. In this research, we will report the enhanced crystalline and piezoelectric properties of (Bi,Na) TiO_3-(Bi,K) TiO_3 ceramics prepared by the two-step sintering process.

KEYWORDS

(Bi,Na) TiO_3-(Bi,K) TiO_3, piezoelectric properties, two-step sintering process

1 | INTRODUCTION

Many different kind of fabrication processes have been tested to enhance the piezoelectric and ferroelectric properties of functional ceramics Such as cold isostatic pressing, hot isostatic pressing, spark plasma sintering, and high energy ball milling process for nano-powders have been proposed and considered as promising methods to enhance the piezoelectric and ferroelectric properties by increasing the density of ceramics.[1–3] However, these fabrication methods are based on advanced processing technologies and quite expensive production process. Therefore, strong demand persists to find alternative methods that improve structural and piezoelectric properties based on the conventional processing technologies. In this research, two-step sintering process will be applied to the (Bi,Na) TiO_3-(Bi,K) TiO_3 ceramics to enhance piezoelectric properties. This process is very simple and does not require extra equipment to employ. It seems that piezoelectric properties of lead-free ceramics can be improved by employing the two-step sintering process due to their densification process. By comparing the two-step sintering process with cold isostatic pressing, piezoelectric charge coefficient of the 0.80 (Bi, Na) TiO_3-0.20 (Bi,K) TiO_3 ceramics was improved. The piezoelectric charge coefficient of 0.80 (Bi,Na) TiO_3-0.20 (Bi,K) TiO_3 ceramics prepared by two step sintering process is 170 pC/N, while that of cold isostatic pressing is around 160 pC/N.[4] In general, the sintering process performed near the temperature around the solidus and liquidus lines. If the sintering process performed too close to the liquidus line, then evaporation can be happened. The two-step sintering process can avoid the evaporation of

volatile components in ceramics.[5] When the piezoelectric composites contain volatile components, such as Na, K and Bi, they can be easily evaporated and lose their structural properties. As a result, electrical properties including piezoelectric properties can be degraded.[6] Therefore, it is very important to find the low temperature sintering process based on the conventional sintering methods, which can improve electrical properties without losing any components during the sintering process. It is reported that the two-step sintering process can effectively improve the electric and piezoelectric properties of lead-free piezoelectric ceramics as the low temperature sintering process.[7–10]

Therefore, as an alternative method to lower the sintering temperature instead of adding ceramic dopants or glass frits, two-step sintering process will be discussed. The two-step sintering process has the strong merit of improving the dielectric and piezoelectric properties through improvement of densification. In addition, the two-step sintering process has advantage of avoiding the evaporation of volatile elements in the piezoelectric ceramics. During the first-step of sintering process, the uniform microstructure can be developed. On the while during the second step of sintering process, it helps to form the enlarged grain size of piezoelectric ceramic by providing driving force for grain growth because second sintering process can provide much longer sintering time compared with that of first sintering process.

The piezoelectric and ferroelectric of sintered ceramics can be enhanced by employing two-step sintering processes by employing densification process. Additionally, the two-step sintering process could reduce the volatilization of substances that possess a low-melting-point.

2 | EXPERIMENTAL

The 0.80 $Bi_{0.5}Na_{0.5}TiO_3$-0.20$Bi_{0.5}K_{0.5}TiO_3$ ceramics were fabricated using the conventional mixed oxide method. The raw materials, such as bismuth oxide (Bi_2O_3, 99.9%), sodium carbonate (Na_2CO_3, 99.0%), potassium carbonate (K_2CO_3, 99.5%), and titanium oxide (TiO_2, 99.9%) powders, were weighed in stoichiometric quantities and ball-milled with zirconium balls (10 mm in diameter) for 24 hours. After drying, the mixture was calcined at 850°C for 2 hours. The calcined powders were mixed with a binder (polyvinyl alcohol) and, using a pressure of 2 metric tons, pressed into discs 12 mm in diameter and about 1 mm in thickness. The discs were sintered by the two-step sintering process shown in Figure 1. The two-step sintering process was operated following several steps. First, the samples were heated at 600°C for burn out PVA for 1 hour with a heating rate of 3°C/min. Then, the samples were reheated from 600°C to 1180°C with a heating rate of 5°C/min. The conventional sintering temperature would typically be held at 1180°C for 2 hours, but for the two-step process this was instead held at 1180°C for 30 minutes, and then decreased to 1080°C at a rate of 5°C/min. The temperature was fixed at 1080°C for various holding times (2, 4, 8, 12, 16, 20, 24, 28, and 32 hours). Silver paste was applied for the electrodes on both sides. The samples were poled at 80°C in silicone oil by applying a direct current (DC) electric field of 4 kV/mm for 15 minutes. The crystalline properties of the $(1-x)$ $Bi_{0.5}Na_{0.5}TiO_3$-$xBi_{0.5}K_{0.5}TiO_3$ (hereafter $(1-x)$ BNT-xBKT) ceramics were then analyzed by X-ray diffraction (XRD, Bruker-AXS; New D8-Advance). The microstructure of the $(1-x)$ BNT-xBKT ceramics was observed by field emission scanning electron microscopy (FE-SEM, Carl Zeiss; SIGMA). The piezoelectric charge constant (d_{33}) was measured using a Berlin-court quasi-static meter and capacitance vs frequency (C-f) was obtained employing an impedance analyzer (Agilent, 4294A) to obtain the dielectric constant (ε_r) and the electromechanical coupling factor (k_p).

3 | RESULTS AND DISCUSSION

Figure 1 shows the sketch of conventional sintering and two-step sintering. In the conventional sintering process, the evaporation can be happened because the sintering process perform near the temperature around the solidus and liquidus lines. Thus, the stoichiometric composition of the ceramics can be mismatched. However, the two-step sintering process, as the low temperature sintering process, can avoid the evaporation of volatile components, such as Bi, Na, and K. As decreasing the porosity of the piezoelectric ceramics, the crystalline property and densification are

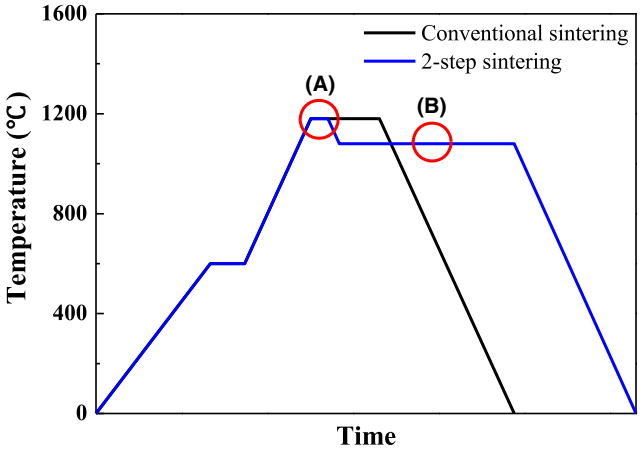

FIGURE 1 The sketch of conventional sintering and two-step sintering (A) holding time: 30 minutes. (B) various holding time (2, 4, 8, 12, 16, 20, 24, 28, 32 h) [Color figure can be viewed at wileyonlinelibrary.com]

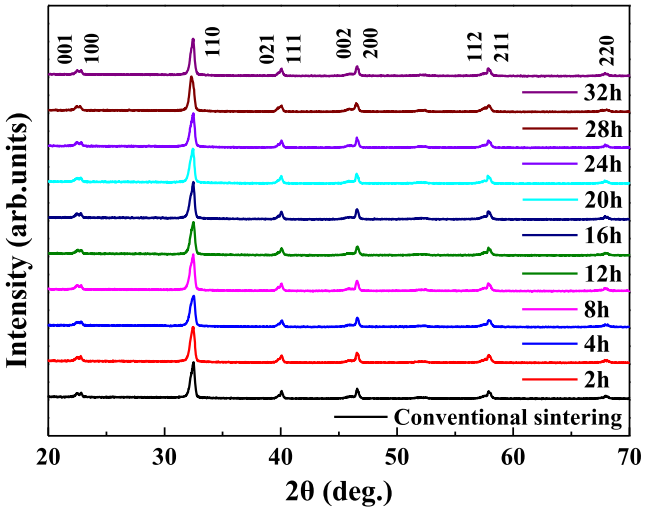

FIGURE 2 The X-ray diffraction (XRD) patterns of the 0.80 (Bi,Na) TiO_3-0.20 (Bi,K) TiO_3 ceramics in conventional sintering and two-step sintering of various holding time (2, 4, 8, 12, 16, 20, 24, 28, 32 h) [Color figure can be viewed at wileyonlinelibrary.com]

improved, which were discussed in Figure 3 and 6. Also, the two-step sintering process has the strong merit of improving the dielectric and piezoelectric properties through improvement of densification and crystalline property.

The XRD patterns obtained from 0.80 BNT-0.20 BKT ceramics with Cu-Kα radiation were shown in Figure 2. Figure 2 also displays the XRD patterns with 2θ ranging from 20 to 70°. Each of the ceramics exhibited a single perovskite phase and no secondary phase was observed. In previous work, the 0.80 BNT-0.20 BKT ceramics was indicated a rhombohedral-tetragonal mixed phase. The rhombohedral structure of pure BNT can be characterized by the splitting of the (021) and (111) peaks around 39° and the (202) single peak near 46.5°. However, the tetragonal structure of BKT is characterized by a (111) single peak near 46.5° and (002) and (200) peaks splitting around 46.5°. In all samples, the splitting of the (021) and (111) peaks were observed around 2θ = 39°. On the other hand, when x = 0.18, 0.20, and 0.30, the splitting of the (002) and (200) peaks were observed at 2θ = 46.5°. Therefore, the 0.80 BNT-0.20 BKT ceramics consist of both tetragonal and rhombohedral phases in conventional sintering and the two-step sintering of various holding times.

Figure 3 shows the peak ratio at (00l) of 0.80 BNT-0.20 BKT ceramics in various holding times (ref., 2, 4, 8, 12, 16, 20, 24, 28, 32). The peak ratio of (00l) was calculated using the following equation.[11]

$$\text{Peak ratio}\,(00l) = \frac{\text{Peak Inensity}\,(00l)}{\sum \text{Peak Intensity}\,(hkl)} \quad (1)$$

Peak ratio (00l) means relative peak intensities of the (00l) orientation, and higher peak ratios represent highly oriented crystalline properties. The peak ratio (00l) was increased at holding times ranging from 2 to 24 hours. This means that crystallites grown for (00l) orientation were improved by increasing the holding time. When the sintering holding time is 24 hours, the peak ratio at (00l) is higher than that of the conventional sintering process. This means that two-step sintering process could improve the (00l) orientation. However, when the sintering holding time is 28 and 32 hours, respectively, the 0.80 BNT-0.20 BKT ceramics have a low peak ratio. Hence, we may conclude that the (00l) orientation was decreased by increasing the holding time.

Figure 4 displays the lattice parameters a and c, and tetragonality c/a of the 0.80 (Bi,Na) TiO_3-0.20 (Bi,K) TiO_3 ceramics sintered conventional and two-step sintering process for various holding times. The lattice parameters were

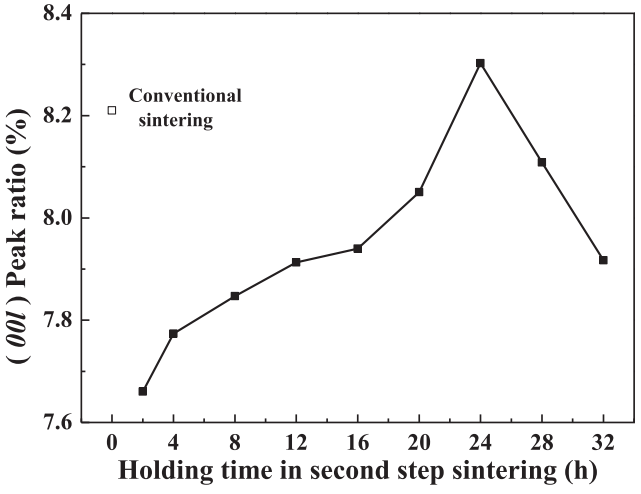

FIGURE 3 Peak ratio at (00l) of the 0.80 (Bi,Na) TiO_3-0.20 (Bi,K) TiO_3 ceramics

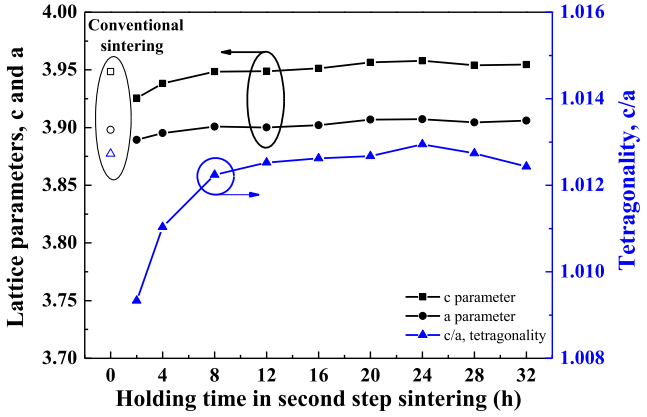

FIGURE 4 Lattice parameters a and c, and tetragonality c/a of the 0.80 (Bi,Na) TiO_3-0.20 (Bi,K) TiO_3 ceramics depending on the holding time in the second step sintering process [Color figure can be viewed at wileyonlinelibrary.com]

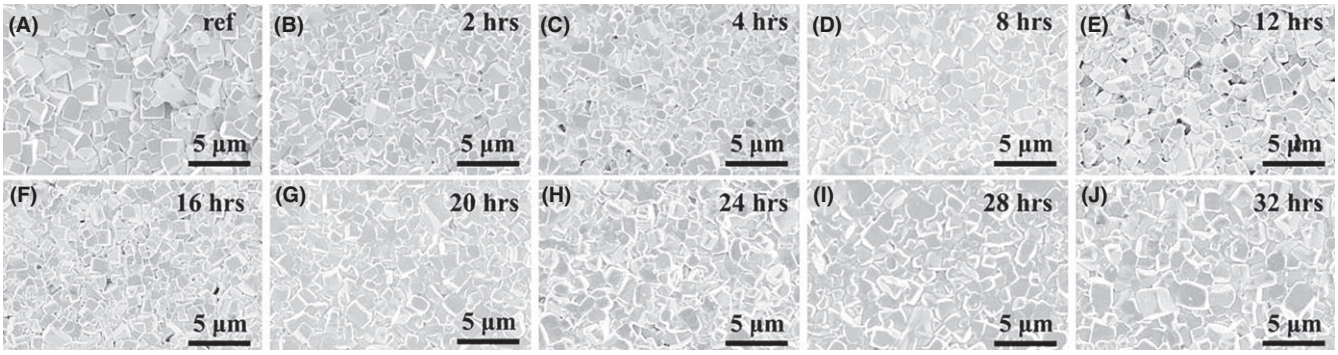

FIGURE 5 Field emission scanning electron microscopy (FE-SEM) images of the 0.80 (Bi,Na) TiO_3-0.20 (Bi,K) TiO_3 ceramics with (A) conventional sintering process and two-step sintering process with various holding time, (B) 2 h (C) 4 h, (D) 8 h, (E) 12 h, (F) 16 h, (G) 24 h, and (H) 32 h

calculated from the XRD patterns (Figure 2) employing Bragg's law and fitted by the Nelson-Riley extrapolation function with the least mean squares method.[12]

$$\frac{C_{\cos\theta} - C_0}{C_0} = A \cdot \cos^2\theta \left(\frac{1}{\sin\theta} + \frac{1}{\theta}\right) \quad (2)$$

where $C_{\cos\theta}$ is the interplane distance calculated from the Bragg peak position at 2θ and A is a constant fitting coefficient. When the samples were sintered using the conventional sintering process, the lattice parameter a and c were 3.89 and 3.94 Å, respectively. The maximum lattice parameter a and c in the two-step sintering process at 24 hours were 3.91 and 3.95 Å, respectively. The tetragonality was calculated from the lattice parameters a and c. The high tetragonality properties appear to be strongly related to high piezoelectric properties.[13] The tetragonality value of samples for the two-step sintering process were increased with increasing holding time. The maximum tetragonality value (c/a) was 1.013 at 24 hours, which is higher than the conventional sintering process. It means that the two-step sintering process with a second step sintering time of 24 hours can produce higher piezoelectric properties than the conventional sintering process.

The microstructure of the 0.80 (Bi,Na) TiO_3-0.20 (Bi,K) TiO_3 piezoelectric ceramics under different sintering conditions are shown in Figure 5. The microstructures of the 0.80 BNT-0.20 BKT ceramic from the conventional sintering process is shown in Figure 5 (A). Figure 5 (a) displays that microstructures of the 0.80 BNT-0.20 BKT ceramic were homogeneous and uniform with grain sizes around 1.5 μm. Figure 5B-H show the specimens that were sintered in the two-step sintering process at various holding times. The observed average grain sizes of the 0.80 BNT-0.20 BKT ceramic with the sintering holding time of 2, 4, 8, 12, 16, 20, 24, 28, and 32 hours were 1.00, 1.07, 1.12, 1.15, 1.20, 1.24, 1.28, 1.35 and 1.48 μm, respectively. The grain sizes of all samples were smaller than when using the conventional sintering process, and as the holding time of the second step sintering process increased, a homogeneous and dense microstructure was observed. By increasing the holding time of the second step sintering process, the grain sizes of the 0.80 BNT-0.20 BKT ceramics were increased. Due to the increased grain size of 0.80 BNT-0.20 BKT ceramics with increased holding time, dielectric and piezoelectric properties were generally increased. However, as increasing the holding time more than 24 hours, the dielectric and piezoelectric properties were decreased due to the decreased densification of 0.80 BNT-0.20 BKT ceramics. It indicates that the increase in the electrical properties of 0.80 BNT-0.20 BKT ceramics is result from the improvement of densification and the elimination of the porosity of specimens. As shown in Figure 6, it confirms that the bulk density of 0.80 BNT-0.20 BKT ceramics increased with holding time up to 24 hours, and then decreased. According to Equation (3) below, the average grain size can be increased by increasing the holding time in the second sintering process.[14]

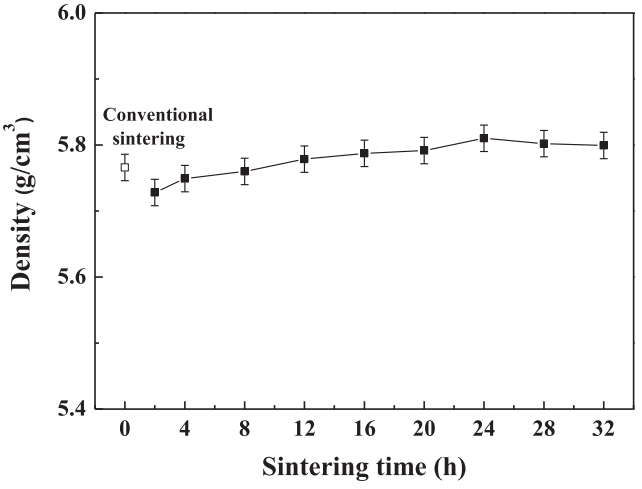

FIGURE 6 Density of the 0.80 (Bi,Na) TiO_3-0.20 (Bi,K) TiO_3 ceramics depending on the holding time in the second step sintering process

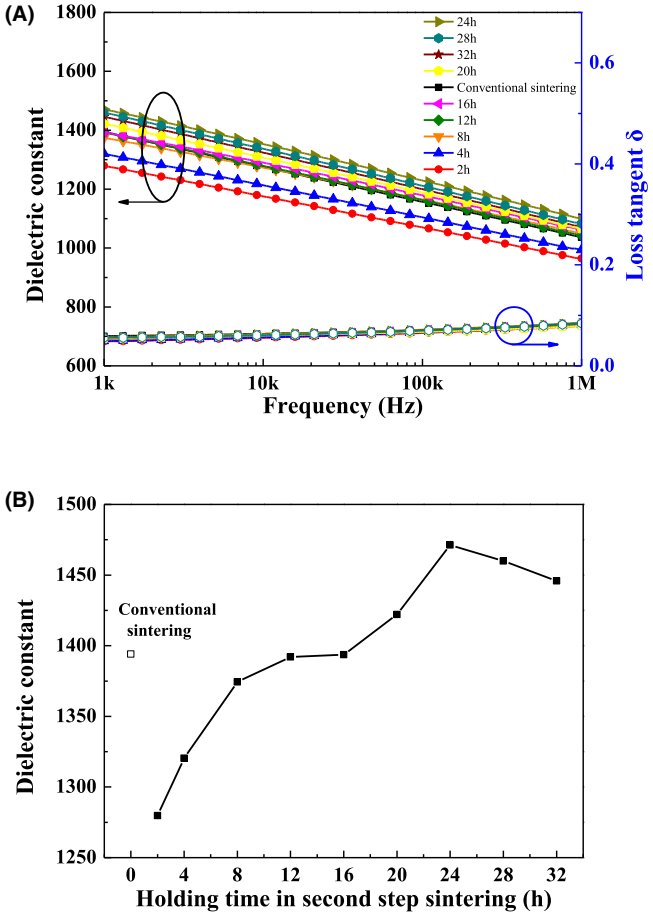

FIGURE 7 Dielectric properties of the 0.80 (Bi,Na) TiO_3-0.20 (Bi,K) TiO_3 ceramics: (A) Frequency dependent dielectric constant and loss tangent and (B) dielectric constant at 1 kHz in the conventional and two-step sintering process depending on the holding time in the second step [Color figure can be viewed at wileyonlinelibrary.com]

$$\log G = \frac{1}{n}\log t + \frac{1}{n}\left(\log \mathbf{K}_0 - 0.434\,\frac{Q}{\mathrm{R}T}\right) \qquad (3)$$

Here, G is the average grain size, n is the kinetic grain growth exponent, t is the sintering time, K_0 is a constant, Q is the apparent activation energy, R is the gas constant and T is the absolute temperature, respectively. As a result, we can confirm that the average grain size can be increased by increasing the sintering temperature and sintering time. Thus, Figure 5B-H indicates similar and smaller grain sizes than the conventional sintering process due to the same maximum temperature and less holding time, respectively. The second step was related to density and enhanced microstructure properties. In this step, the grain growth is limited due to a lower temperature. However, enhanced density was measured by increasing holding time as shown in Figure 6. The second step temperature could influence the driving forces of the grain boundary to enhance densification.[15] Also, it can reduce pores and prevent the volatilization of low melting point materials. As shown in Figure 6, the density of samples was increased by increasing the holding time. The maximum value of the density is 5.81 g/cm^3 and the density of the sample from conventional sintering process was 5.76 g/cm^3. Hence, the two-sintering process enhanced microstructure properties.

Figure 7 illustrates dielectric properties of the 0.80 BNT-0.20 BKT in the conventional sintering process and the two-step sintering process with various holding times. Figure 7A displays the frequency dependent dielectric constant (ε_r) and loss tangent δ of the 0.80 BNT-0.20 BKT in a frequency range from 1 kHz to 1 MHz. The dielectric constant of all sintered samples was decreased with increased frequency, and the loss tangent was increased with increasing frequency. This behavior is associated with the effect of dipoles.[16] The direction of the polarization can be switched due to alignment under an applied AC electric field. In addition, increments of frequency occurred with decreased polarization. As shown in Figure 7B, the dielectric constants of samples were increased as holding time increased up to 24 hours. The maximum of the dielectric constant is 1471.29 at 24 hours, which is higher than the constants arising from the conventional sintering process. This high value of the dielectric constant probably come from the highest (00*l*) peak ratio of 24 hours compared with other holding times.

As shown in Figure 8, the values of the piezoelectric charge constant (d_{33}) and the electromechanical coupling factor (k_p) of the 0.80 BNT-0.20 BKT ceramics with the two-step sintering process were increased when holding time was increased from 2 to 24 hours, and then subsequently decreased. This behavior is well coincided with that of the (00*l*) peak ratio, which was discussed in

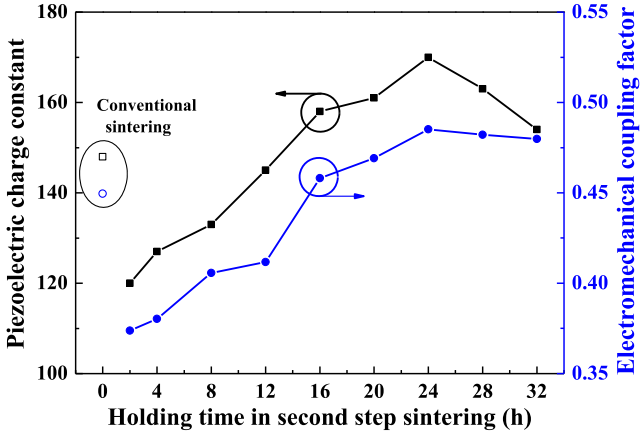

FIGURE 8 The piezoelectric charge constant (d_{33}) and the electromechanical coupling factor (k_p) of the 0.80 (Bi,Na) TiO_3-0.20 (Bi,K) TiO_3 ceramics prepared in a conventional and two-step sintering process depending on the holding time in the second step [Color figure can be viewed at wileyonlinelibrary.com]

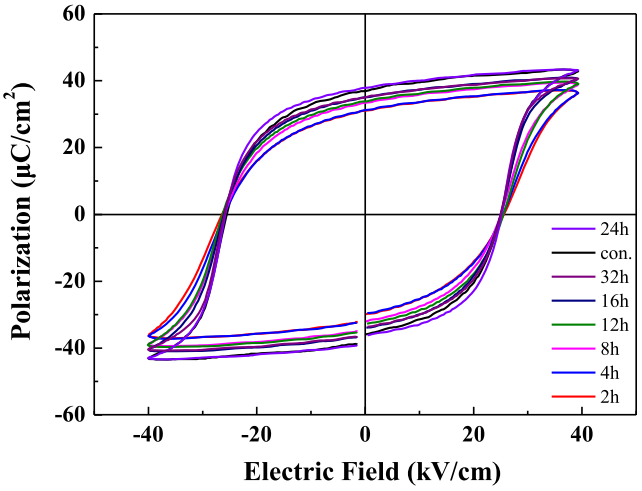

FIGURE 9 Hysteresis loops of the 0.80 (Bi,Na) TiO_3-0.20 (Bi,K) TiO_3 prepared in a conventional and two-step sintering process depending on the holding time in the second step at 0.1 Hz [Color figure can be viewed at wileyonlinelibrary.com]

Figure 3. As we have previously discussed, the (00*l*) crystalline properties were related with the piezoelectric constant and electromechanical coupling properties. Also, these excellent piezoelectric properties can be associated with the microstructure of the material, as discussed in Figure 5. In general, the two-step sintering process has the strong merit of improvement of the densification. Therefore, it seems that the increase in the piezoelectric properties of 0.80 BNT-0.20 BKT ceramics were resulted from the improvement of densification and the elimination of the porosity of specimens. For various specimens within the two-step sintering process, the d_{33} and k_p values varied from 120 to 170 pC/N and 0.37 to 0.48, respectively. The maximum values of d_{33} of 170 pC/N and k_p of 0.48 were achieved for the 0.80 BNT-0.2 BKT specimens at 24 hours. These are higher than the sample which was produced by the conventional sintering process.

Figure 9 shows the polarization vs electric field (P-E) hysteresis loop of 0.80 (Bi,Na) TiO_3-0.20 (Bi,K) TiO_3 ceramics prepared in a conventional and two-step sintering process depending on the holding time in the second step. The P-E hysteresis loops were measured under an applied electric field of 40 kV/cm at 0.1 Hz. The P-E hysteresis loops of all specimens were saturation. When holding time is increased, the P-E hysteresis loops became slim which is increased remnant polarization (P_r). The maximum value of remnant polarization was 37.87 μC/cm^2 and the minimum value of coercive field was 24 kV/cm at 24 hours.

Figure 10 shows the piezoelectric properties of the 0.80 (Bi,Na) TiO_3-0.20 (Bi,K) TiO_3 ceramics with holding time of 24 hours based on the poling condition. Figure 10A displays poling temperature dependent piezoelectric charge constant with fixed poling electric field of 40 kV/cm. As

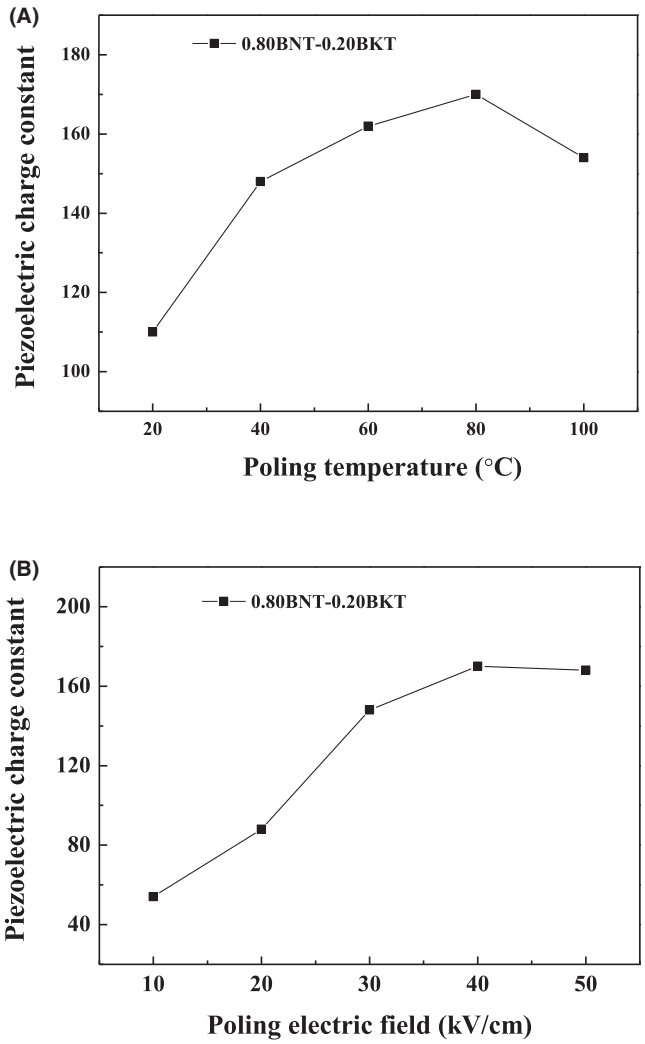

FIGURE 10 The piezoelectric properties of the 0.80 (Bi,Na) TiO_3-0.20 (Bi,K) TiO_3 ceramics with holding time of 24 h: (A) The poling temperature dependent piezoelectric charge constant, (B) The poling electric field dependent piezoelectric charge constant

shown in figure, the piezoelectric charge constant increased with poling temperature up to 80°C, and then decreased. It seems that there are strong relationship between the piezoelectric charge coefficient and poling condition.[17] Figure 10B shows poling electric field dependent piezoelectric charge constant with fixed poling temperature of 80°C. The piezoelectric charge constant reached maximum value of 170 pC/N when poled in 40 kV/cm. As a result, it means that the piezoelectric properties of the 0.80 (Bi,Na) TiO_3-0.20 (Bi,K) TiO_3 ceramics are significantly affected by the poling temperature and electric field.

4 | CONCLUSIONS

The 0.80 BNT-0.20 BKT lead-free piezoelectric ceramics were fabricated by a two-step sintering process and the conventional process. The XRD patterns showed that the

0.80 BNT-0.20 BKT samples in all conditions show tetragonal and rhombohedral structures. Increasing the first-step sintering temperature increased grain size, while increasing the holding time in the second step sintering process caused the grain to become dense. Also, as the holding time was increased, the (00*l*) peak ratio of the 0.80 BNT-0.20 BKT ceramics increased. The two-step sintered specimens with a holding time of 24 hours in the second step sintering process showed uniform grain size and produced the maximum values of density, ε_r, d_{33} and k_p, which were 5.81 g/cm^3, 1471.29, 170 pC/N and 0.48, respectively. While conventionally sintered, 0.80BNT-0.20BKT ceramics have ε_r, d_{33} and k_p of 1394.06, 148 pC/N and 0.44, respectively. Therefore, by comparing the piezoelectric coefficient d_{33} of 0.80 BNT-0.20 BKT, two-step sintered sample with 24 hours of holding time in the second step sintering process have the piezoelectric properties that were enhanced by 13.9% when compared with conventionally sintered ceramics.

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REFERENCES

1. Mahesh MLV, Bhanu Prasad VV, James AR. Enhanced dielectric and ferroelectric properties of lead-free $Ba(Zr_{0.15}Ti_{0.85})O_3$ ceramics compacted by cold isostatic pressing. *J Alloy Compd* 2014;611:43-49.
2. Li JF, Wang K, Zhang BP, Zhang LM. Ferroelectric and Piezoelectric Properties of Fine-Grained $Na_{0.5}K_{0.5}NbO_3$ Lead-Free Piezoelectric Ceramics Prepared by Spark Plasma Sintering. *J Am Ceram Soc*. 2006;89:706-709.
3. Wang S, Li JF, Watanabe R, Esashi M. Fabrication of Lead Zirconate Titanate Microrods for1–3 Piezocomposites Using Hot Isostatic Pressing with Silicon Molds. *J Am Ceram Soc*. 1999;82:213-215.
4. Yoshii K, Hiruma Y, Nagata H, Takenaka T. Electrical Properties and Depolarization Temperature of $(Bi_{1/2}Na_{1/2})TiO_3$–$(Bi_{1/2}K_{1/2})TiO_3$ Lead-free Piezoelectric Ceramics. *Jpn J Appl Phys*. 2006;45:4493-4496.
5. Hao J, Bai W, Shen B, Zhai J. Improved piezoelectric properties of $(K_xNa_{1-x})_{0.94}Li_{0.06}NbO_3$ lead-free ceramics fabricated by combining two-step sintering. *J Alloy Compd* 2012;534:13-19.
6. Lee Y, Yoo J, Lee K, Kim I, Song J, Park YW. Dielectric and piezoelectric characteristics of the non-stoichiometric (Na, K) NbO3 ceramics doped with CuO. *J Alloy Compd*. 2010;506:872-876.
7. Zheng T, Wu J. Enhanced piezoelectricity over a wide sintering temperature (400-1050°C) range in potassium sodium niobate-based ceramics by two step sintering. *J Mater Chem A*. 2015;3:6722-6780.
8. Wu J, Wang Y. Two-step sintering of new potassium sodium niobate ceramics: a high d_{33} and wide sintering temperature range. *Dalton Trans*. 2014;43:12836-12841.
9. Wu J, Xiao D, Zhu J. Potassium−Sodium Niobate Lead-Free Piezoelectric Materials: Past, Present, and Future of Phase Boundaries. *Chem Rev*. 2015;115:2559-2595.
10. Manotham S, Kruea-In C, Rujijangagul G. Properties of $0.94Bi_{0.5}Na_{0.5}TiO_3$–$0.06BiAlO_3$ Ceramics Prepared by Two Steps Sintering Technique. *Ferroelectrics* 2014;458:152-157.
11. Balzar D, Ledbetter H, Stephens PW, Park ET, Routbort JL. Dislocation-density changes upon poling of polycrystalline $BaTiO_3$. *Phys Rev B*. 1990;59:3414-3420.
12. Kang WS, Koh JH. $(1-x)Bi_{0.5}Na_{0.5}TiO_3$–$xBaTiO_3$ lead-free piezoelectric ceramics for energy-harvesting applications. *J Eur Ceram Soc*. 2015;35:2057-2064.
13. Zhang S, Randall CA, Shrout TR. Dielectric, piezoelectric and elastic properties of tetragonal $BiScO_3$-$PbTiO_3$ single crystal with single domain. *Solid State Commun*. 2004;131:41-45.
14. Li HB, Li Y, Wang DW, Lu R, Yuan J, Cao MS. Effects of ZnO nanoneedles addition on the mechanical and piezoelectric properties of hard PZT-based composites. *J Mater Sci*. 2013;24:1463-1468.
15. Karaki T, Yan K, Adachi M. Barium Titanate Piezoelectric Ceramics Manufactured by Two-Step Sintering. *Jpn J Appl Phys*. 2007;46:7035-7038.
16. Kang WS, Lee SK, Koh JH. AC conductivity and dielectric properties of (Bi, Na)TiO3–BaTiO3 lead free ceramics. *Ceram Int*. 2015;41:6925-6932.
17. Zhang B, Wu J, Wu B, Xiao D, Zhu J. Effects of sintering temperature and poling conditions on the electrical properties of $Bi_{0.50}(Na_{0.70}K_{0.20}Li_{0.10})_{0.50}TiO_3$ piezoelectric ceramics. *J Alloy Compd* 2012;525:53-57.

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