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Origin of thermal depolarization in piezoelectric ceramics

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ABSTRACT

PZT, BSPT, PT, and two lead-free Bi_2WO_6, as five of the most representative piezoelectric systems with different Curie points were selected to study the origin of thermal depolarization in piezoelectric ceramics. The temperature dependences of dielectric loss and dielectric constant curves show that dielectric loss peaks of PZT_{MN}, $BSPT_{PNW}$, PT_{BFM}, BW_{Li}, and BW_{La} are at 330 °C, 392.2 °C, 495 °C, 598.8 °C, and 781 °C, respectively. The results of thermal depoling measurement show that the depolarization temperature is closely related to the dielectric loss peak. And the peak points of dielectric loss curves below or at the Curie temperature are the points of complete depolarization.

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1. Introduction

Piezoelectric ceramics are ideal for most kinds of electromechanical transducers. Each piezoelectric ceramic has a particular operating limit for temperature, voltage, and stress. And the limits are determined by the chemical composition. The operation of the material out of the limitations will cause partial or total depolarization. Usually, the temperature limitations gain more attentions than the voltage and stress limitations for piezoelectric ceramics. As the operating temperature increases, piezoelectric performance of a material decreases, until complete and permanent depolarization occurs at the Curie temperature. In practice, the operating temperature limit which usually was named as depolarization temperature actually is commonly below the Curie point [1–6]. The depolarization temperature is the most important reference for the practical use of piezoelectric ceramic. However, the study about the origin of depolarization is not as much as that on the Curie point. M. Zhu et al. [3] reported that the depolarization temperature was influenced by the lattice distortion other than the oxygen vacancies. S. Zhang et al. [1] pointed out that the depolarizing was the result of an R–T ferroelectric phase transition. Most attentions were preferred to focus on the crystal structure or phase transition. In order to improve the study on depolarizing of piezoelectric ceramic, five of the most representative samples about $Pb(Ti,Zr)O_3$ (PZT, T_c is around 320 °C) [7], $BiScO_3$–$PbTiO_3$(BSPT, T_c is around 450 °C) [8], $PbTiO_3$ (PT, T_c is around 490 °C) [9], and Bi_2WO_6 (BW, T_c is around 900 °C) [10] with different Curie points were selected. In this paper, for the sake of reducing the influence factor as much as possible, single phase samples were prepared. The origin of thermal depolarizing for piezoelectric ceramic was discussed.

2. Experimental procedures and methods

The research samples with formula of $0.94Pb_{0.98}Sr_{0.02}Ti_{0.48}Zr_{0.52}O_3$-$0.06Pb(Mn_{1/3}Nb_{2/3})O_3$ (after abbreviated as PZT_{MN}), $0.608BiScO_3$-$0.342PbTiO_3$-$0.05Pb(Zn_{1/3}Nb_{2/3})O_3$ (abbreviated as $BSPT_{PZN}$), $0.608BiScO_3$-$0.342PbTiO_3$-$0.05Pb(Ni_{1/2}W_{1/2})O_3$ (abbreviated as $BSPT_{PNW}$), $0.95PbTiO_3$-$0.05\ Bi_2FeMnO_6$ (abbreviated as PT_{BFM}), $0.95Bi_2WO_6$-$0.05Li_2CO_3$ (abbreviated as BW_{Li}), and $0.90Bi_2WO_6$-$0.10La_2O_3$ (abbreviated as BW_{La}) were prepared by conventional solid-state reaction techniques using high purity (better than 99.9%) Pb_3O_4, ZrO_2, Nb_2O_5, TiO_2, ZnO, Sc_2O_3, Bi_2O_3, NiO, WO_6, Fe_2O_3, $MnCO_3$, La_2O_3, and Li_2CO_3 as the starting materials. PZT_{MN}, $BSPT_{PZN}$, $BSPT_{PNW}$ and PT_{BFM} were prepared by two-step procedure [11]. In the first step, $Pb_{0.98}Sr_{0.02}Ti_{0.48}Zr_{0.52}O_3$, $0.608BiScO_3$-$0.342PbTiO_3$, $PbTiO_3$, $Pb(Mn_{1/3}Nb_{2/3})O_3$, $Pb(Zn_{1/3}Nb_{2/3})O_3$, $Pb(Ni_{1/2}W_{1/2})O_3$, Bi_2FeMnO_6, and Bi_2WO_6 were milled using de-ionized water and zirconia balls for 6 h, and the dried powders were calcined at 760 °C, 850 °C, 670 °C,760 °C, 760 °C, 760 °C, and 760 °C for 2.5 h, respectively. The calcined powders were mixed following the formula of PZT_{MN}, $BSPT_{PZN}$, $BSPT_{PNW}$, PT_{BFM}, BW_{Li}, BW_{La}, and then pressed into pellets with 12 mm in diameter and 1 mm in thickness with of appropriate amount PVA (5%) added. The PZT_{MN} ceramics were sintered at 1250 °C for 4 h. The $BSPT_{PZN}$ and $BSPT_{PNW}$ ceramics were sintered at 1150 °C for 4 h. The PT_{BFM} ceramics were sintered at 850 °C for 4 h. The BW_{Li} and BW_{La} ceramics were sintered at 850 °C for 4 h. The powder X-ray diffraction analysis of PZT_{MN}, $BSPT_{PZN}$, $BSPT_{PNW}$, and PT_{BFM} was carried out at the 1W1A beamline at the Beijing synchrotron radiation facility (BSRF) with an X-ray wave length of 0.1547 nm. The powder X-ray diffraction analysis

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of BW_{Li} and BW_{La} was carried out at the 4B9A beamline at the Beijing synchrotron radiation facility (BSRF) with an X-ray wave length of 0.15406 nm. The crystal structure of all samples was determined by *ab initio* and Rietveld refinement [12]. The ceramic pellets for electrical characterization were polished and coated with silver electrodes, and then PZT_{MN}, $BSPT_{PZN}$, $BSPT_{PNW}$ were poled at ~120 °C for 20 min under an electric field of ~4 kV/mm, PT_{BFM} were poled at ~160 °C for 20 min under an electric field of ~4 kV/mm, BW_{Li} and BW_{La} were poled at ~70 °C for 20 min under an electric field of ~4 kV/mm in a silicone oil bath, respectively. The piezoelectric properties were measured after 24 h aging at room temperature. Temperature dependence of dielectric properties were measured using a computer controlled TH2826 (Changzhou, China) high frequency LCR meter from room temperature to 550–850 °C. The planar electromechanical coupling factor k_p and Q_m were derived by the resonance and anti-resonance method. The piezoelectric constant d_{33} was measured by a quasistatic piezoelectric ZJ-3D d_{33} meter produced by Institute of Acoustics, Chinese Academy of Sciences. Thermal depoling experiments were conducted by annealing the poled samples at various temperatures for 240 h, and d_{33} values were measured after cooling down to room temperature.

3. Results and discussion

Fig. 1 shows the synchrotron radiation patterns of PZT_{MN} (Fig. 1a), $BSPT_{PNW}$ (Fig. 1b), PT_{BFM} (Fig. 1c), BW_{Li} (Fig. 1d), and BW_{La} (Fig. 1e) ceramics. All samples show single phase. In order to determine the crystal structure of each sample, *ab initio* and Rietveld refinement [12] were adopted. The *ab-initio* calculation results from DICVOL [13] show that the symmetry group of PZT_{MN}, $BSPT_{PNW}$, PT_{BFM}, BW_{Li}, and BW_{La} are Pmm2 (25), Pmm2 (25), Pmm2 (25), Aba2 (41), and Aba2 (41), respectively. After refinement without atom site via Fullprof [12], the atom sites were calculated by Fourier maps calculation. The results were refinement again via Fullprof, and the final structure parameters were shown in Table 1. Part of visual calculation results were shown in supplement.

Fig. 2 shows the temperature dependences of dielectric loss (right Y) and dielectric constant (left Y) of the PZT_{MN} (Fig. 2a), $BSPT_{PNW}$ (Fig. 2b), PT_{BFM} (Fig. 2c), BW_{Li} (Fig. 2d), and BW_{La} (Fig. 2e) ceramics. The dielectric loss peaks of PZT_{MN}, $BSPT_{PNW}$, PT_{BFM}, BW_{Li}, and BW_{La} locate at 330 °C, 392 °C, 495 °C, 600 °C, and 781 °C, respectively. The dielectric constant peaks of PZT_{MN}, $BSPT_{PNW}$, and PT_{BFM} are 330 °C, 402 °C, and 506 °C, respectively. The dielectric constant peaks of both BW_{Li}, and BW_{La} are above 850 °C. The dielectric constant peak is well known as Curie temperature which is related to the transition from ferroelectric to paraelectric phase. According to this theory, the depolarization occurred at the Curie temperature when the ferroelectricity disappeared. However, most researches show that the depolarization temperature is below or far below the Curie temperature [1–6]. The origin of depolarization is important to determine the depolarization temperature. In order to exclude the complex influence factors of

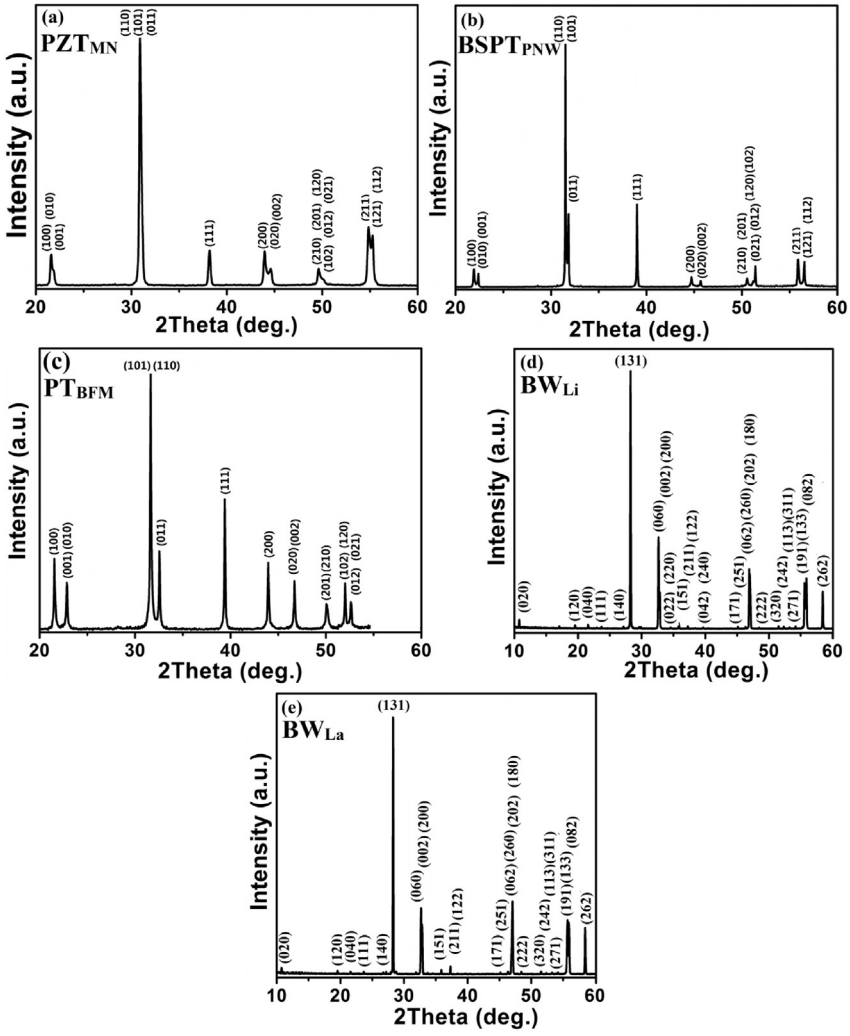

Fig. 1. The synchrotron radiation patterns of PZT_{MN} (a), $BSPT_{PNW}$ (b), PT_{BFM} (c), BW_{Li} (d), BW_{La} (e) ceramics.

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