

## Supplemental Materials

# Temperature Independence of piezoelectric properties for High-Performance BiFeO<sub>3</sub>-BaTiO<sub>3</sub> Lead-free Piezoelectric Ceramics up to 300°C

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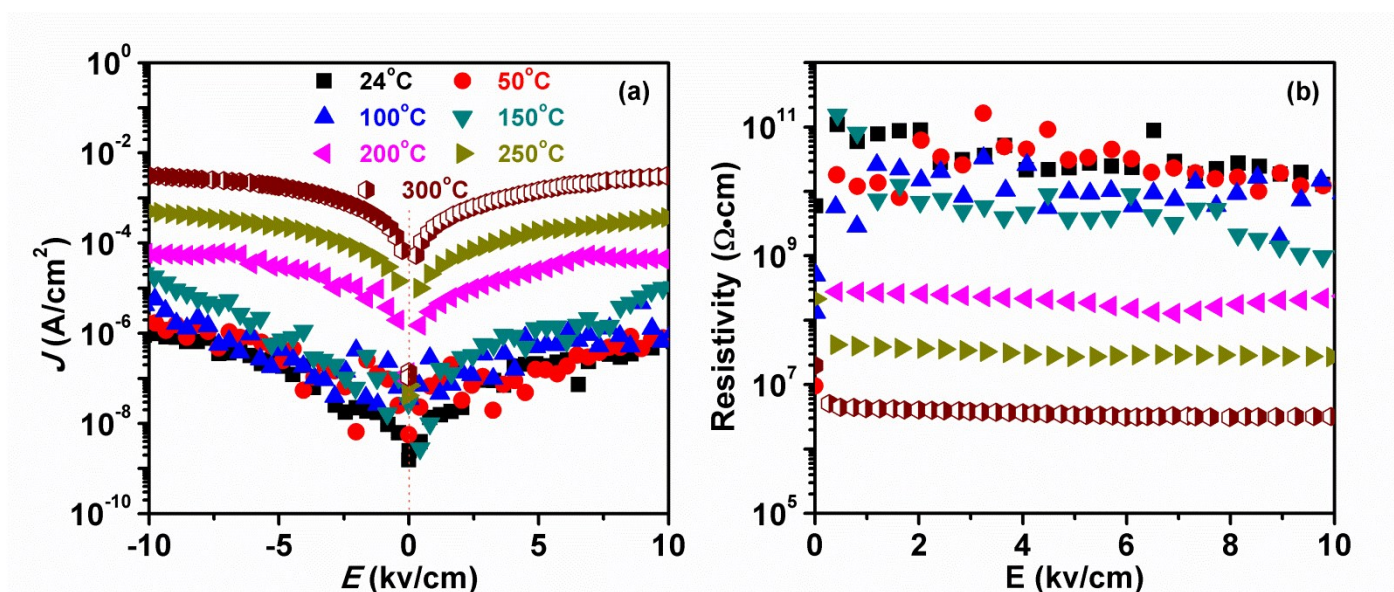


Fig.S1 Temperature dependence of leakage current (a) and resistivity (b) for BF-0.3BT ceramic

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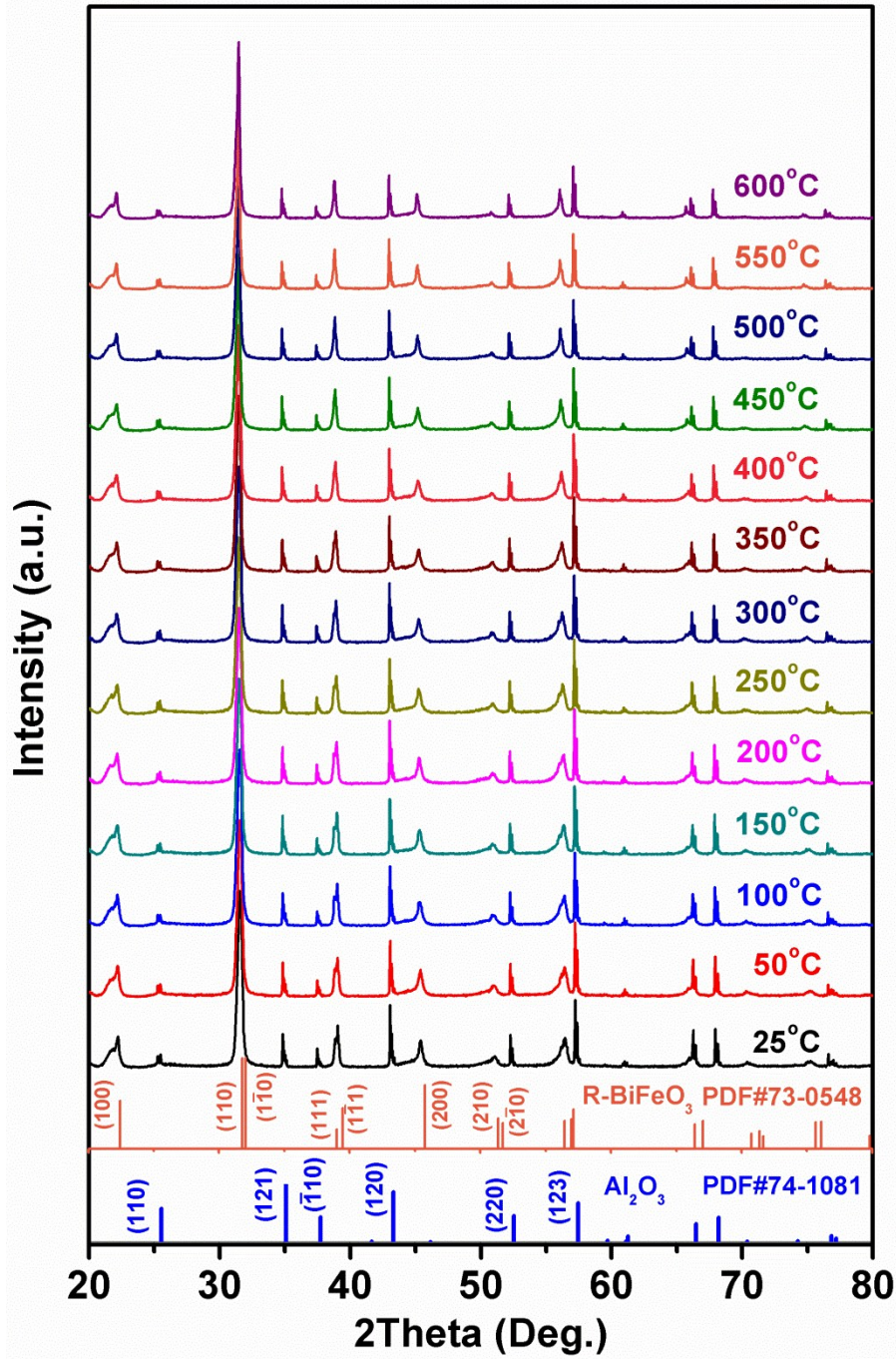


Fig S2 Temperature dependence of X-ray diffraction patterns for BF-0.3BT ceramics in a selected  $2\theta$  range of  $20^\circ$ - $80^\circ$ , the  $\text{Al}_2\text{O}_3$  diffraction peaks are detected in all samples, which are caused by crucible.

The sample exhibit a perovskite structure without any trace of impurity phase within the detectable limit of the XRD as shown in Fig. S3. The enlarged XRD pattern in inset of Fig.S3 shows two diffraction peaks around  $22^\circ$ , indicating the lattice parameters are  $a=b \neq c$ , which correspond to *T* phase. Thus, the phase structure of samples consists of the *R* and *T* two phases, rather than single *R* phase. This phenomenon has been verified in some papers [1, 2].

[1]. M. H. Lee, D. J. Kim, J. S. Park, S. W. Kim, T. K. Song, M. H. Kim, W. J. Kim, D. Do and I. K. Jeong, “High-Performance Lead-Free Piezoceramics with High Curie Temperatures,” *Adv. Mater.* **27**, 6976-6982 (2015).

[2]. L. F. Zhu, B. P. Zhang, Z. C. Zhang, S. Li, L. J. Wang and L. J. Zheng, “Piezoelectric, ferroelectric and ferromagnetic properties of  $(1-x)\text{BiFeO}_3\text{-}x\text{BaTiO}_3$  lead-free ceramics near morphotropic phase boundary,” *J. Mater. Sci. Mater. Electron.* **29**(3), 2307-2315 (2018)

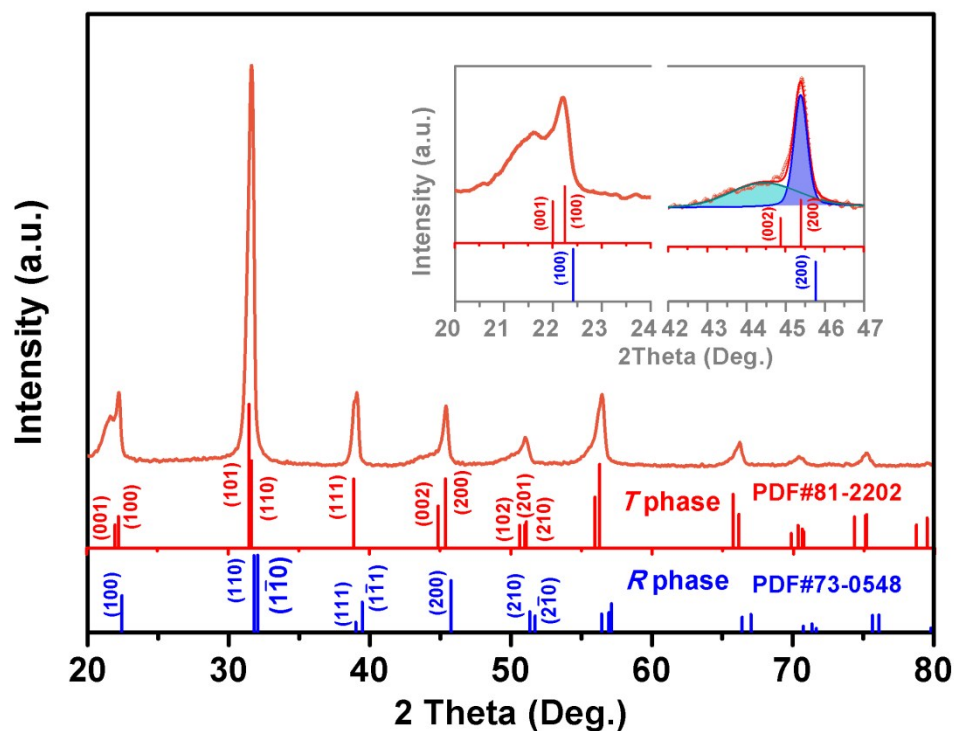


Fig. S3 X-ray diffraction patterns for BF-0.3BT ceramics in a selected  $2\theta$  range of  $20^\circ$ - $80^\circ$  measured at room temperature.

The standard diffraction peaks cited from  $\text{BiFeO}_3$  (BF) with *R* symmetry (PDF#73-0548) and  $\text{BaTiO}_3$  (BT) with *T* phase (PDF#81-2202) are indicated by vertical lines for comparison.

Fig.S4 shows minor strain-field hysteresis loops were used to calculate the complex and imaginary piezoelectric coefficients. Following the equations (S1-S3), the real part of  $d_{33}$  ( $d'$ ) was calculated. In addition, the extrinsic contribution ratio for BF-0.3BT ceramics was also evaluated as following the equation (S4 and S5).

$$d'' = A_{EM} / \pi E_0^2 \quad (S1)$$

$$d^* = \chi_{p-p} / 2E_0 \quad (S2)$$

$$d' = \sqrt{d^{*2} - d''^2} \quad (S3)$$

$$d' = d_{init} + a_d E_0 \quad (S4)$$

$$Extrinsic(\%) = \frac{d_{extrinsic}}{(d_{intrinsic} + d_{extrinsic})} \times 100\% = \frac{a_d E_0}{(d_{init} + a_d E_0)} \times 100\% \quad (S5)$$

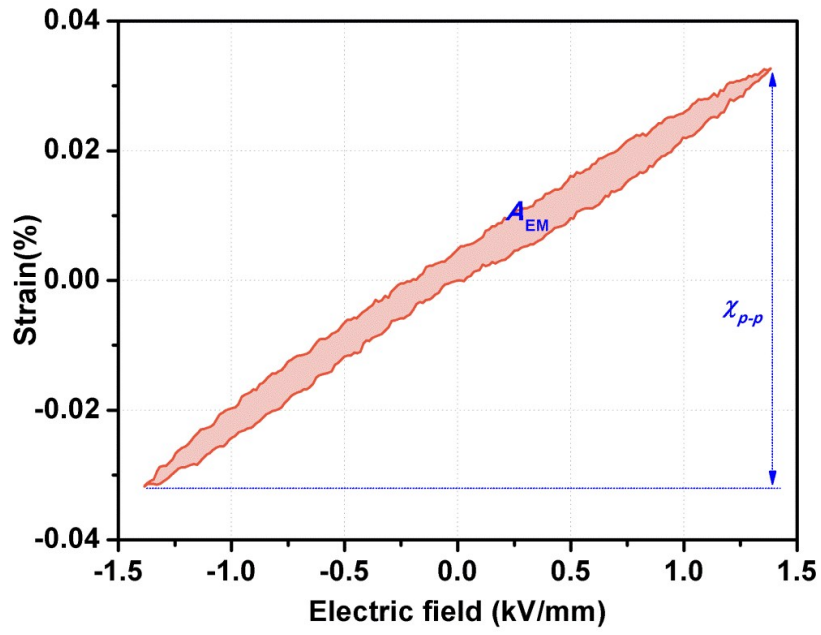


Fig. S4 (Color online) Minor strain-field hysteresis loops were used to calculate the complex and imaginary piezoelectric coefficients. A typical minor hysteresis loop is presented (BF-0.3BT), measured at 5 Hz and 1.4kV/cm



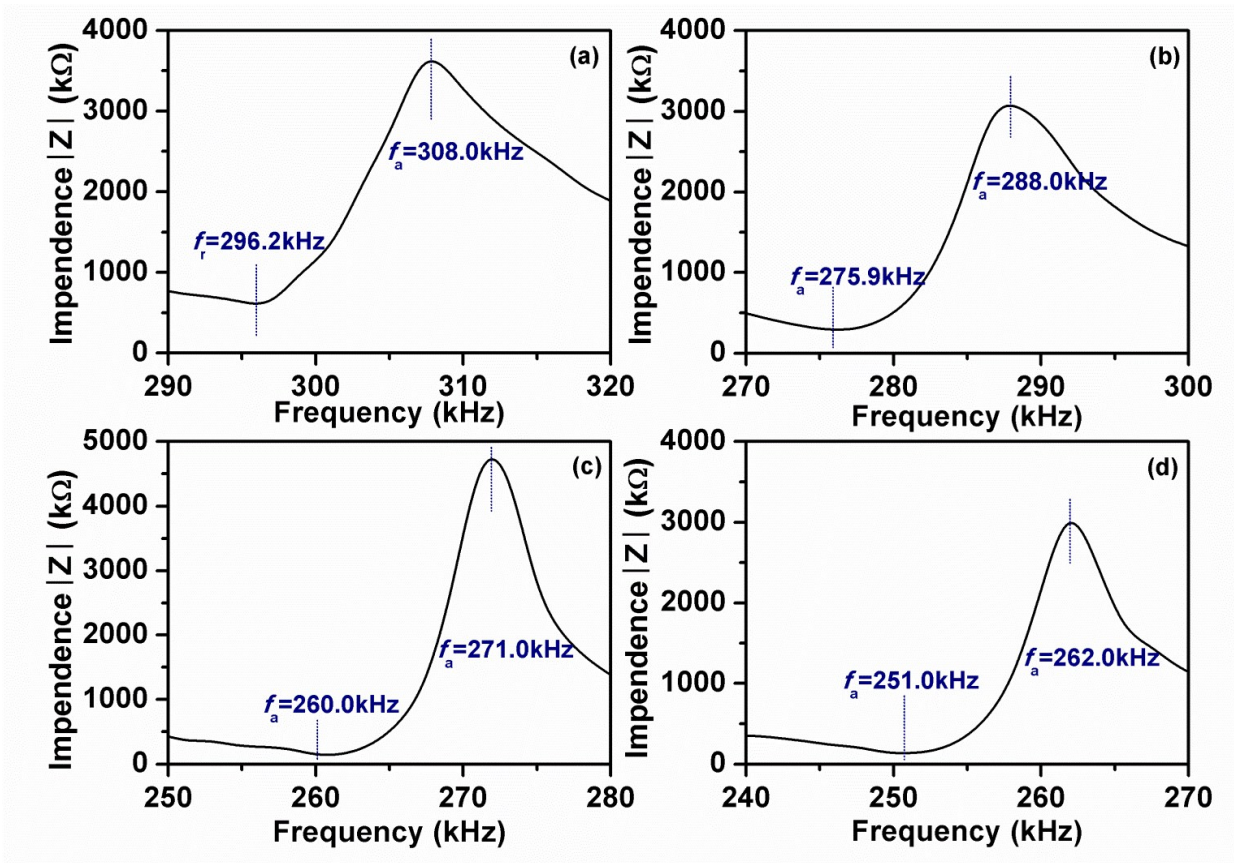


Fig. S5 The impedance spectra  $|Z|$  as a function of frequency of BF-0.3BT ceramics with different measured temperature, (a) 30 °C, (b) 100 °C, (c) 200 °C and (d) 300 °C

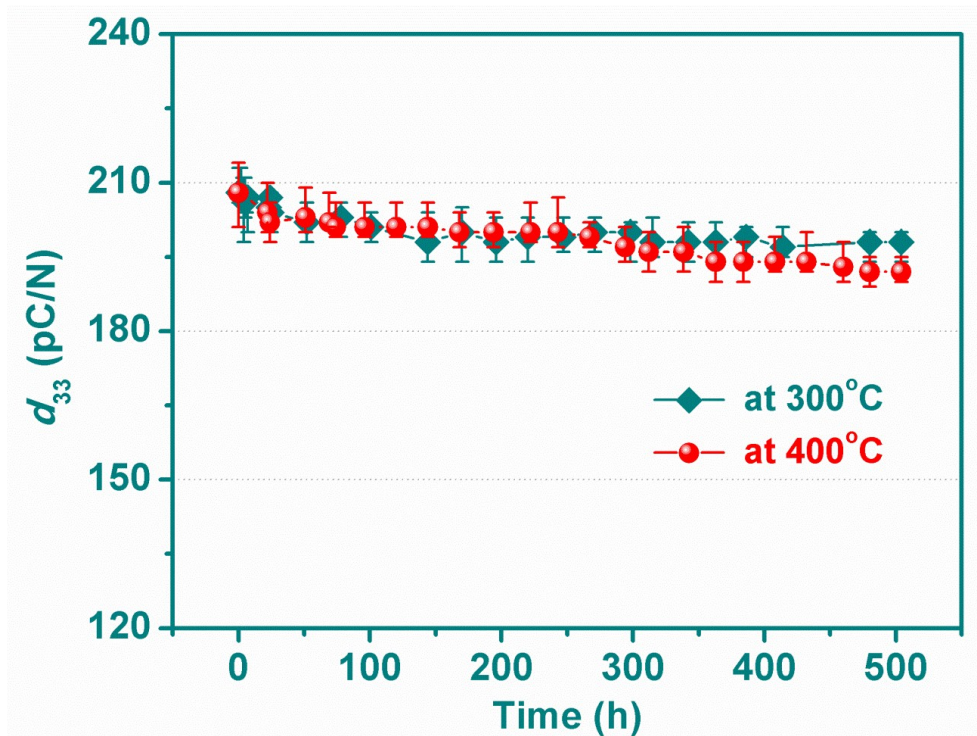


Fig. S6 Aging of piezoelectric coefficient ( $d_{33}$ ) of BF-0.3BT ceramics was measured after heat treatment at 300 °C and 400