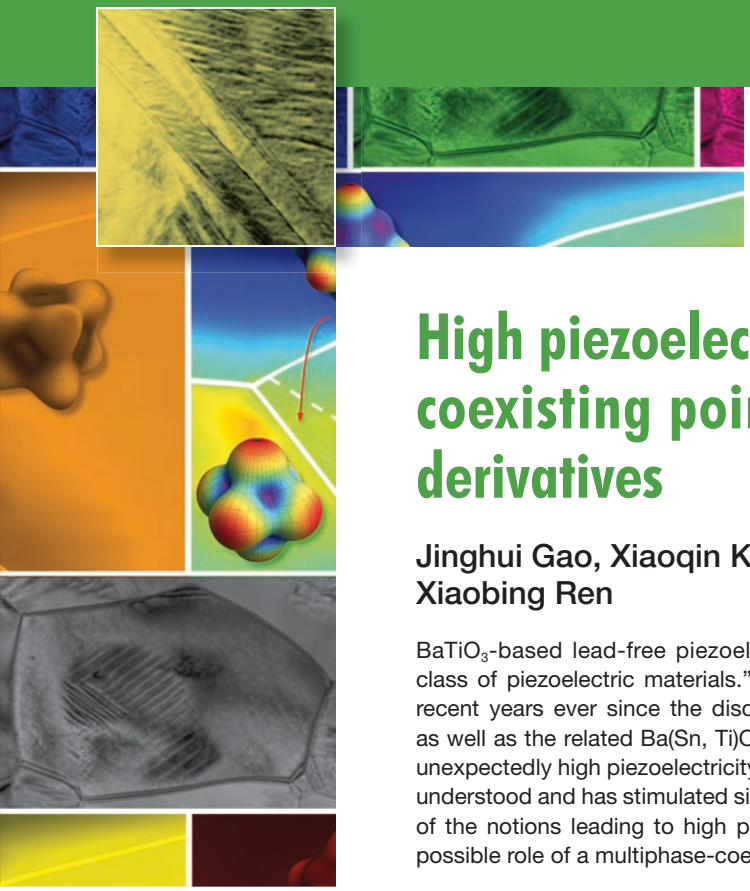


High piezoelectricity by multiphase coexisting point: Barium titanate derivatives

Jinghui Gao, Xiaoqin Ke, Matias Acosta, Julia Glaum, and Xiaobing Ren

$BaTiO_3$-based lead-free piezoelectric materials have long been known as "a mediocre class of piezoelectric materials." However, they have seen significant renewed interest in recent years ever since the discovery of high piezoelectricity in Ba(Zr, Ti)O_3-(Ba, Ca)TiO_3 as well as the related Ba(Sn, Ti)O_3-(Ba, Ca)TiO_3 and Ba(Hf, Ti)O_3-(Ba, Ca)TiO_3 systems. The unexpectedly high piezoelectricity in this class of $BaTiO_3$ (BT)-based materials is still not well understood and has stimulated significant research activity. We present a concise discussion of the notions leading to high piezoelectricity in $BaTiO_3$-based systems. In particular, the possible role of a multiphase-coexisting point is highlighted.

Introduction

Barium titanate ($BaTiO_3$ [BT]) was discovered in the 1940s during World War II.[1] Before the emergence of Pb(Zr,Ti)O_3 (PZT) in the late 1950s, BT had been the most important known piezoelectric material due to its piezoelectric coefficient (induced polarization per unit stress applied to the material) d_{33} = 120–190 pC/N, which is significantly higher than that of older piezoelectric materials such as triglycine sulfate (TGS), potassium dihydrogen phosphate (KDP), and quartz. However, compared to PZT, which shows outstanding piezoelectricity with d_{33} = 300–600 pC/N, BT and BT-based materials appeared to be "mediocre" in terms of both piezoelectricity and T_C (Curie temperature, approximately 130°C). Thus, they were soon replaced by PZT in piezoelectric applications.[2] It should be noted that although pure BT can show higher piezoelectricity in some special forms such as single crystals,[3,4] textured polycrystals,[5] or fine-grain/domain ceramics,[6,7] it is challenging to achieve PZT-like, intrinsically high piezoelectricity in simple chemically modified BT-based ceramics.

The Pb-free surge over the past 20 years has revived significant research on the piezoelectricity of BT-based materials, in an attempt to discover a BT-based ceramic that is comparable to PZT in piezoelectric characteristics.[3–9] In 2009, Liu and Ren reported large piezoelectric response for the Ba(Zr,Ti)O_3-(Ba,Ca)TiO_3 (BZT-BCT) ceramic system with a d_{33} coefficient larger than 600 pC/N, which is comparable to soft PZT ceramics with high sensitivity and permittivity.[10] These ceramics are made by a standard solid-state reaction method and are random polycrystals, suggesting that the high piezoelectricity stems from its inherent effect. Further studies have shown that similar values of high piezoelectricity are also present in a generic class of BT-based materials, including Ba(Sn,Ti)O_3-(Ba,Ca)TiO_3 (BTS-BCT) and Ba(Hf,Ti)O_3-(Ba,Ca)TiO_3 (BHT-BCT).[11,12] In contrast to the mediocre piezoelectricity of BT-based ceramics reported over the past 60 years, such soft-PZT-like piezoelectricity in BT-based ceramics came as a surprise to the piezoelectric community, as it can be a potential replacement for Pb-based piezoelectric materials. Significant efforts have thus been made toward understanding and explaining this phenomenon.[13–29] In the following, we provide a brief review of the state-of-the-art key results and our current understanding. A more comprehensive review can be found in Reference 30.

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Generic phase diagram of $BaTiO_3$-based systems

High-performance $BaTiO_3$-based systems (BZT-BCT, BST-BCT, BHT-BCT) exhibit similar phase diagrams, as shown in **Figure 1**a–c.[10–12] They all exhibit a PZT-like phase diagram characterized by a morphotropic phase boundary (MPB) (shown in Figure 1) separating two ferroelectric phases (tetragonal [T] and rhombohedral [R]).[10,31–34] As highlighted in Figure 1a–c, the most important feature of the phase diagrams in this generic class of materials is that MPB starts from a multiphase coexisting point (MCP) in the phase diagrams, where cubic (C), tetragonal (T), and rhombohedral (R) phases meet. We will show how the MCP results in important consequences that account for the high piezoelectricity of this new class of BT-based materials.

More detailed recent investigations suggest the existence of a bridging orthorhombic (O) phase in BZT-BCT between the T and R phases at MPB such that the MPB is not a line, but an extended O phase[35–38] (Figure 1d). It is likely that the bridging O phase also exists in BTS-BCT and BHT-BCT systems and further studies need to verify this hypothesis.

Interestingly, the research development in BZT-BCT is similar to that of PZT, where MPB was initially reported to be a line separating the R and T phases, but later turned out to be a bridging monoclinic phase.[39] The existence of bridging O phase at the MPB suggests that the MCP is not a C-T-R triple point, but a C-T-O-R multiphase coexisting point. It should be noted that there is some uncertainty as to whether the MCP is a point or an extended region.[13,14] Further research is needed to clarify this.

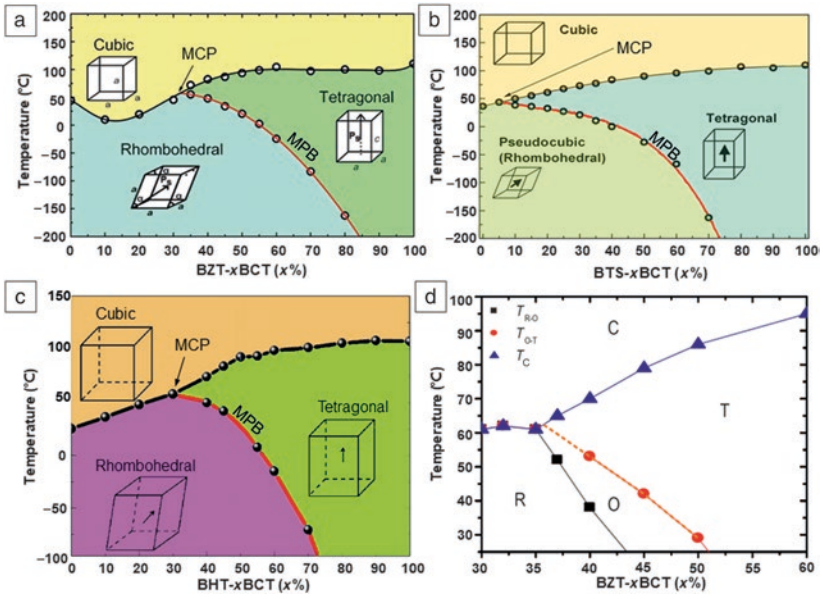


Figure 1. Generic phase diagram of $BaTiO_3$-based systems. (a) Phase diagram of $Ba(Zr,Ti)O_3$-$(Ba,Ca)TiO_3$ (BZT-BCT).[10] (b) Phase diagram of $Ba(Sn, Ti)O_3$-$(Ba,Ca)TiO_3$ (BTS-BCT)[11] and (c) $Ba(Hf,Ti)O_3$-$(Ba,Ca)TiO_3$ (BHT-BCT).[12] (d) Modified phase diagram of BZT-BCT,[13] which amends the Liu–Ren phase diagram by adding an intermediate orthorhombic phase (O). Note: MCP, multiphase coexisting point; MPB, morphotropic phase boundary; R, rhombohedral; T, tetragonal; C, cubic; T_{R-O}, transition temperature of R to O phase; T_{O-T}, transition temperature of O to T phase; T_C, Curie temperature; $\mathbf{P}_s$, spontaneous polarization.

Properties in relation to MPB and MCP

Figure 2 shows an example in BZT-BCT of ferroelectricity, dielectric permittivity, and more importantly, piezoelectricity, showing maximum values around the MPB composition;[10] similar results have been found for BTS-BCT and BHT-BCT.[11,12] In these systems, the optimal d_{33} at the MPB can reach 500–620 pC/N, comparable to that of soft PZT. It is also found that the permittivity peaks at the MCP, with $\varepsilon_r \sim 25{,}000$,[10] which is much higher than the value for pure BT ceramic (~10,000).[40] This suggests that the MCP is the most field-susceptible location in the phase diagram.

Since BT and BT-based ceramics normally do not show high piezoelectricity, the anomalously high piezoelectricity observed for BZT-BCT generic systems needs an explanation. Much effort has been made in an attempt to answer this important question.[19–29] Factors such as elastic softening,[13,14,16–18] high domain wall mobility,[19,20] phase instability,[15,21] and degree of poling[14,22] have been considered, but no single factor is considered deterministic. The role of these factors in improving the piezoelectric performance has been summarized in recent reviews.[30,41] In the following, we show that the high piezoelectricity of the BZT-BCT generic systems stems from the MCP in the system, and this explains why normal BT and other BT-based systems do not show impressive piezoelectricity (in a random polycrystalline form).

Tricriticality and microstructural anomaly at the MCP

Figure 3a shows that the MCP is not merely a point where C, T, R, and O phases meet; it is a tricritical point (i.e., a point where the first-order transition crosses over into a second-order transition), as characterized by vanishing transition hysteresis and latent heat.[10,42] This means that at the MCP, no energy barrier exists among the four phases; thus this situation corresponds to a flat or frying-pan-shaped free-energy landscape,[10] as shown in Figure 3b(i–ii). The MPB, starting from the MCP, partially inherits this low anisotropy feature and thus generates high d_{33} at the MPB composition. This makes a difference between the BZT-BCT generic systems and other BT-based systems and thus provides an explanation for the unexpectedly high piezoelectricity in such systems.

Figure 4 shows that the tricriticality of the MCP yields interesting microstructural features[40–44] not seen in standard BT-based materials. At the MCP (the tricritical point), mottled nanodomains appear with blurred boundaries and miniaturized domains (Figure 4b),[40,42]

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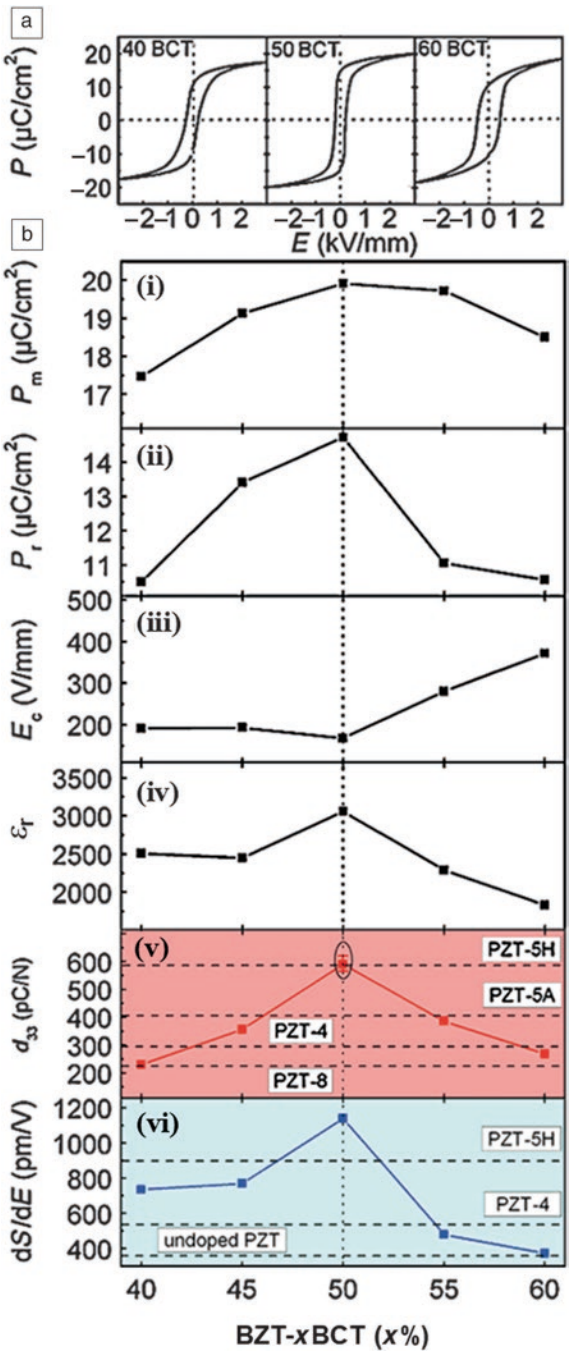


Figure 2. Ferroeletricity, dielectric permittivity, and piezoelectricity of (1-*x*%)$Ba(Zr_{0.2}Ti_{0.8})O_3$-*x*%$(Ba_{0.7}Ca_{0.3})TiO_3$ (BZT-*x*BCT) around the morphotropic phase boundary (MPB) composition.[10] (a) (Left to right) Hysteresis loops of BZT-40 BCT, BZT-50 BCT, and BZT-60 BCT, respectively. (b[i]) Saturation polarization P_m, (b[ii]) Remnant polarization P_r, (b[iii]) coercive field E_c, (b[iv]) permittivity ε_r, (b[v]) piezoelectric coefficients d_{33}, and (b[vi]) converse piezoelectric coefficient d*S*/d*E*. These coefficients show maximums at the MPB composition (dashed lines in the figures indicate the value of d_{33} or d*S*/d*E* of PZT-5H, PZT-5A, PZT-4, and PZT-8, which are commercialized PZT ceramics that differ in piezoelectric properties, depending on their specific applications) Note: *P*, polarization, *S*, strain; *E*, electric field.

contrasting with large regular domains in the off-MCP compositions. The isotropic nanodomains at the MCP are consistent with the vanishing polarization anisotropy or vanishing energy barrier.[42] Such a microstructure is an indication of the flat free-energy landscape shown in Figure 3. The off-MCP compositions show a hierarchical domain structure at the MPB in BZT-BCT,[33,34,45–49] which indicates low but nonvanishing polarization anisotropy[10] (as shown in Figure 3c[i–ii]). Therefore, the high piezoelectric performance at the MPB seems to stem from the vanishing polarization anisotropy at MCP.

Modeling

Much effort has been made to understand the observed experimental phenomena using Landau–Devonshire-type phenomenological models,[50–52] in which the free energy of the system can be expressed in the form of a polynomial with respect to a so-called order parameter that is the spontaneous polarization. However, to date, there is no predictive theory that can explain why BZT-BCT generic systems show a tricritical MCP and high piezoelectricity, whereas normal BT-based materials do not. This is at the heart of the problem and awaits future theoretical efforts.

Summary

We have briefly reviewed the phase diagrams, piezoelectric properties, microstructure, and theoretical modeling of BZT-BCT-type generic systems. The role of the multiphase coexisting point (MCP) and the MPB region starting from a MCP in enhancing the piezoelectric performance is highlighted. A better understanding of the observed high piezoelectricity in the hitherto "mediocre" BT-based piezoelectric materials may be transferable to the understanding and development of high-performance Pb-free piezoelectric materials in other systems.[53,54]

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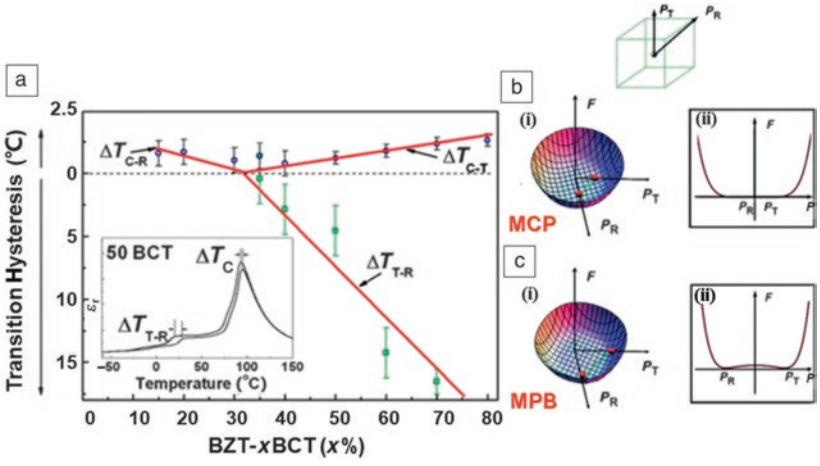
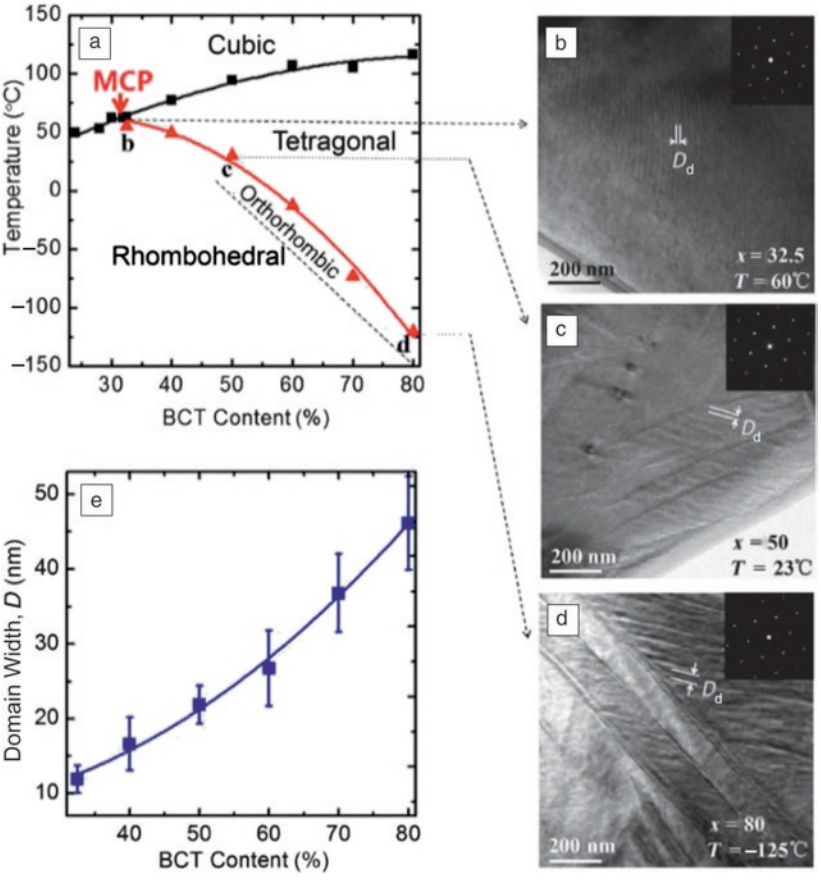


Figure 3. Tricritical behavior of (1-*x*%)Ba($Zr_{0.2}Ti_{0.8}$)O_3-*x*%($Ba_{0.7}Ca_{0.3}$)TiO_3 (BZT-*x*BCT) ceramic and the associated free-energy landscape. (a) Thermal hysteresis of the three transitions (C-T, C-R, and T-R) for BZT-*x*BCT ceramic nearly vanishes when approaching the multiphase coexisting point (MCP), and the inset shows measurement of thermal hysteresis for BZT-50BCT using heating and cooling permittivity–temperature (ε_r–*T*) curves. (b[i–ii]) MCP tricritical point showing flat free-energy landscape between P_T and P_R. (c[i–ii]) Morphotropic phase boundary (MPB) composition at room temperature showing low, but nonvanishing, free-energy landscape between P_T and P_R.[10] Note; C, cubic; T, tetragonal; R, rhombohedral; PZT, Pb(Zr, Ti)O_3; P_T, axis for tetragonal polarization; P_R, axis for rhombohedral polarization; ΔT_C, thermal hysteresis for Curie temperature; $\Delta T_{T\text{-}R}$, thermal hysteresis for T-R transition temperature (indicated by green data); $\Delta T_{C\text{-}T}$, thermal hysteresis for C-T transition temperature (indicated by blue data); $\Delta T_{C\text{-}R}$, thermal hysteresis for C-R transition temperature (indicated by blue data); *F*, free energy.



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Figure 4. Transmission electron microscope (TEM) observations for domain structures of (1-*x*%)Ba($Zr_{0.2}Ti_{0.8}$)O_3-*x*%($Ba_{0.7}Ca_{0.3}$)TiO_3 (BZT-*x*BCT) on a morphotropic phase boundary (MPB) line approaching the multiphase coexisting point (MCP) tricritical point.[42] (a) Phase diagram for BZT-BCT (($Ba_{0.7}Ca_{0.3}$)TiO_3) showing the locations (points b, c, and d) of the TEM observations. (b) At MCP tricritical point, mottled nanodomains appear with blurred boundaries. (c–d) Off-MCP compositions show hierarchical domain structure at MPB in BZT-*x*BCT at (c) *x* = 50% and (d) *x* = 80%. The diffraction patterns in (b–d) indicate that all images were viewed from the same crystallographic orientation. (e) Domain width decreases on approaching MCP. Note: *T*, temperature; D_d, domain width.

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