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High temperature blocking force measurements of soft lead zirconate titanate

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Abstract

Piezoelectric actuators outperform other technological solutions in the area of high speed, high force, and high accuracy displacement, but are only able to generate strains of about 0.2%. The load capability is generally quantified in terms of a blocking force, which is the force sustained under electric field at zero displacement. Stress-strain curves in a temperature regime from room temperature until 150°C on electrically loaded soft lead zirconate titanate (PZT) are generated to determine the blocking stress. The ensuing non-linear behavior is discussed in terms of ferroelectric and ferroelastic switching and contrasted to idealized linear constitutive behavior as often assumed by manufacturers. The blocking stress is shown to increase with temperature due to an additional stiffening effect as a function of electric field. The actual mechanical work done is found to be larger than in the idealized case where linear constitutive behavior is assumed.

1. Introduction

Strong electromechanical coupling in ferroelectric materials, i.e., lead zirconate titanate (PZT), makes them ideal candidates for actuator applications, such as diesel fuel injection systems [1, 2]. Typical piezoelectric actuators consist of a cofired multilayer design, where the internal electrodes terminate abruptly within the bulk of the actuator and are used to electrically activate thin, stacked layers of piezoelectric ceramic material [3]. Variation in the thickness of the layers can reduce the required actuation voltage while maintaining the necessary maximum electric field, which is typically on the order of 2 to 3 kV mm⁻¹ for doped PZT [4]. Multilayer actuators, currently being implemented into diesel injection systems, have a number of advantages over existing electromagnetic solenoid systems, such as short actuation time and greater fuel mass injection precision, resulting in increased fuel efficiency, lower engine noise, and decreased CO₂ emissions [1, 5]. However, in addition to elevated ambient temperatures found in a diesel engine, piezoactuators can also experience “self-heating” due to the loss factor of donor-doped PZT (soft PZT) [6, 7]. Acceptor-doped PZT (hard PZT) displays lower losses as well as lower piezoelectric coefficients, reducing both the self-heat generation and the overall strain that can be obtained from a piezoactuator. Previous studies have shown temperature increases for multilayer stack actuators of PZT on the order of 60 to

100 °C, depending on actuator size, actuation frequency, actuation time, and duty cycle [6-10], which can impact actuator characteristics.

Actuator applications have a number of stringent requirements, including actuation response time, stroke amplitude, and blocking force. The free displacement (D_f) and the blocking force (F_b) are common values supplied by the manufacturer for specific piezoactuator models. Because displacement and force are measured, not strain and stress, comparison between different commercially available actuators with different materials, geometries, and insulating materials (e.g., end cap material and insulating lacquer) can be difficult. Blocking stress and free strain, however, are material, not application, specific parameters that can be directly compared. Figure 1 shows the typical schematic representation of the force-displacement relationship presented by manufacturers. Most often the free displacement is measured during the application of a specific driving voltage on a poled sample, followed by the application of a monotonically increasing mechanical load until the actuator height is reduced back to the original value of the poled, zero electric field state. The load required to reduce the electric field-induced displacement to zero is termed the blocking force. A line is drawn between these two points, which represents the assumed operational range of this particular actuator. With increasing applied voltage there is an increase in both the free strain and the blocking force.

During the electrical activation of a piezoactuator work is done on a spring load, represented in the inset (Fig. 1) as a linear spring element with a spring constant k , which is equal to the area below the load-displacement curve. For a linear spring load this is one half of the product of the force applied and the distance through which it acts. When the spring constant is zero there is no force required to compress the spring. In this situation the displacement of the actuator is equivalent to the free displacement at activation voltage V , $D_f(V)$, and no work is done. When the spring constant is infinite there is no displacement of the actuator. In this case the force on the actuator is equivalent to the blocking force at activation voltage V , $F_b(V)$, and there is again no work done. However, for realistic loads that lie between these two limits the maximum achievable work occurs at one half of the blocking force and free displacement for the activation voltage used, often termed an impedance matched spring load [11]. The maximum work done for other nonlinear loads, however, does not necessarily occur exactly at one half of the blocking force and free displacement. The linear force-displacement model is obviously highly idealistic; ignoring much of the nonlinear behavior inherent in ferroelectric materials with a large change in Young's modulus during application of compressive stresses [12-14].

Ferroelectric/ferroelastic lead zirconate titanate (PZT) with various dopants has been utilized for actuator applications due to exceptional material behavior over a range of temperatures [15]. However, macroscopic constitutive behavior of PZT in response to electric field [15, 16] and stress [13, 14, 17] has been shown to depend strongly on temperature. A temperature increase results in a decrease in the coercive electric field [16] as well as an observed increase in the maximum strain obtained during unipolar cycling [15], both principally due to an increase in domain wall mobility. However, in addition to their ability to ferroelectrically switch in response to an external electric field, ferroelastic domain switching due to mechanical loads is also possible. In application, actuators usually operate under a compressive prestress to reduce the tensile stresses at electrode edges. This compressive stress enhances back-switching during electrical loading with the effect that multilayer actuators exhibit a maximum in achievable strain at approximately -30 MPa at room temperature [18, 19]. However, at elevated temperatures a modest compressive preload (~5 MPa) can be large enough to cause a loss of the remanent state [13], necessitating an additional poling cycle.

This study presents high temperature ferroelectric and ferroelastic data used to determine the free strain and blocking stress of a commonly utilized material in piezoactuators. To date there have been few studies of high temperature ferroelasticity [13, 14, 17], despite the increasing importance of ferroelectrics in certain automotive applications. To this end, this study focuses on experimentally measuring the high temperature blocking stress of bulk polycrystalline donor-doped PZT commonly found in commercially available multilayer actuators. Specimens are applied with large field electrical and mechanical loads while the longitudinal strain is measured at various temperatures. Results are contrasted with common industry measurement procedures.

2. Experimental Methods

Experimental measurements were conducted on a commercial-grade soft PZT, $\text{Pb}_{0.99}(\text{Zr}_{0.45}\text{Ti}_{0.47}(\text{Ni}_{0.33}\text{Sb}_{0.67})_{0.08})\text{O}_3$ (PIC 151, PI Ceramics, Lederhose, Germany) with a composition in the vicinity of the morphotropic phase boundary. This material was chosen due to the excellent electromechanical properties, i.e., $d_{33} \approx 500$ pC/N, and its wide use in commercially produced actuators. Previous XRD measurements have shown a loss in spontaneous strain between 300 and 320 °C, corresponding with the Curie temperature [16]. Cylindrical samples with a diameter of approximately 5.9 mm and a height of 6 mm were core drilled from a PZT plate for mechanical testing. Samples electrically tested were ground to a height of 3 mm. Prior to electromechanical testing each sample was annealed at 600 °C for 1

hr, with a cooling rate of 50 °C hr⁻¹. Annealing was performed to ensure a fully depoled sample and to help eliminate possible residual stress induced during specimen preparation. Samples were sputtered with an Ag electrode on both circular faces perpendicular to the 6 mm axis.

During this investigation a custom built differential dilatometer setup was used to measure the longitudinal strain of each sample in response to applied electric field and compressive stress (Fig. 2). The experimental arrangement, previously described in detail [13], was fitted with a silicone oil bath enabling the measurement of the stress-strain behavior in response to an applied electric field. The sample was positioned between a tungsten carbide platform and conical loading die, shown in Fig. 2 directly above and below the specimen. Strain was measured as the relative displacement by an LVDT located well outside of the heating chamber. Alumina rods contacted the bottom of the platform and the bottom of the conical loading die, essentially measuring the combined strain of the sample and the platform. Additional experiments were conducted at each temperature under consideration to determine the strain of the platform without a sample. This was used to correct the strain measurements of the PZT samples.

To determine blocking force it was required to apply electrical and mechanical loads simultaneously. A schematic of the strain as a function of the electric field during the testing procedure is shown in Fig. 3. Each specimen was initially unpoled (Point a). During testing the PZT samples were poled at the temperature of interest with an electric field of 3 kV mm⁻¹ for ten minutes (Point b). The nonlinear poling curve is shown in Fig. 3 as the dashed line between Points a and b. Following the poling stage the electric field was removed and the measured displacement was set to zero (Point c). The horizontal dotted line at Point c indicates the reference strain used to determine the blocking force. A constant dc electric field (0.5, 1, 1.5, 2, 2.5, and 3 kV mm⁻¹) was again applied to the already poled sample (Point d), followed by the application of a uniaxial mechanical load (Point e). The strain difference between the poled, electrically unloaded state and the poled, electrically loaded state approximately equated to the free strain of the material, ϵ_f . The mechanical load was applied monotonically to the poled, electrically activated specimen with a loading rate of 1.8 MPa s⁻¹ from an initial stress of -3.7 MPa to a maximum stress of approximately -112 MPa. Electromechanical testing was done at 25, 50, 100, and 150 °C. Each temperature and electric field loading combination was repeated to ensure measurement repeatability.

It was found that at elevated temperatures without the application of an electric field there was significant creep of PZT accelerated by the mechanical preload, resulting in the

time-dependent loss of the remanent state of the poled sample. At lower temperatures (25 and 50 °C) this was not a problem. To alleviate this issue, additional samples were electrically tested in a silicone oil bath to determine the stress-free unipolar electric field-strain behavior as a function of temperature. These measurements were used to calibrate the initial free strain measurements of the mechanical loading experiments. At 25 and 50 °C the additional free strain measurements corresponded well with the free strain determined in the load frame. This will be discussed in further detail in the following sections. Specimens were poled with an electric field of 3 kV mm⁻¹ for ten minutes prior to unipolar electric field cycling, which consisted of five triangular loading cycles with a maximum electric field of 3 kV mm⁻¹ and a frequency of 0.05 Hz. During testing the strain was measured in response to electric field at 25, 50, 100, and 150 °C. A minimum of five samples were tested at each temperature to ensure measurement consistency.

3. Results

Due to the above mentioned creep at elevated temperatures additional quasi-static unipolar electric field-strain measurements were performed. Figure 4 displays representative unipolar hysteresis curves at 25 and 150 °C. From these measurements the free strain was determined at various levels of electric field represented in Fig. 4 as the points on the loading curve. It can be observed that there was an increase in measured free strain at each electric field level (0.5, 1, 1.5, 2, 2.5, and 3 kV mm⁻¹) with an increase in temperature from 25 to 150 °C.

At each of the electric field levels and temperatures under consideration the free strain was determined from the unipolar strain-electric field hysteresis measurements (Fig. 5a). For each electric field level there is an apparent maximum in the achievable strain at 100 °C. This trend can be more clearly observed when the free strain is normalized by the maximum strain at the corresponding electric field value (ϵ_{max}), where it can be seen that the maximum strain occurs at 100 °C for 1, 2, and 3 kV mm⁻¹ (Fig. 5b). Although not shown, this trend was also observed for the other electric fields as well. Below 100 °C there is an obvious increase in ratio of free strain to maximum strain with increased electric field loading. At 150 °C, however, there is a decrease in measured free strain with respect to the maximum strain for each electric field loading level.

The ferroelastic constitutive behavior in response to a constant electric field was characterized as a function of temperature. Figure 6 displays representative stress-strain measurements for various electric field levels at 25 °C. It can be clearly observed that with increasing electric field there is a corresponding increase in both the free strain (strain at zero

stress) and the blocking stress (stress at zero strain). In addition, the measured stress-strain behavior is nonlinear and depends on the applied electric field and temperature. Due to the limitations of the measurement setup the free strain does not begin from zero stress. This represents a known error. The tangent modulus was measured at the initial stress (-3.7 MPa) and used to approximate the material behavior at low loads. The point where the dotted line crossed the x -axis was assumed to be the free strain.

4. Discussion

The electrical activation of ferroelectric materials has both an intrinsic and extrinsic component. During the initial application of an electric field on an unpoled specimen there is large scale domain switching (extrinsic) at the coercive field resulting in nonlinear strain-electric field behavior observed during the poling process, shown schematically in Fig. 3 (dashed line a-b), in addition to intrinsic contributions. However, during the determination of the blocking stress the remanent strain is taken as the reference strain. This neglects the large strain gains from initial, large-scale domain reorientation during the determination of the stress required to bring the strain back to zero (Fig. 3). This is because piezoactuators, where blocking stress is an important characteristic, are actuated in the poled state by a unipolar electric field. It can be observed from Fig. 5 that there is an increase in the free strain up to 100 °C, followed by a decrease at 150 °C. Previous experimental measurements have shown an increase in poling strain with temperature for various compositions of LaSr-doped PZT around the morphotropic phase boundary (MPB) [15]. This increase in poling strain, however, is not linear and was shown to saturate, dependent on PZ/PT ratio and dopant content. This is due, in part, to the shape of the MPB [20], which curves towards the rhombohedral phase. As the temperature is increased there can be a shift in phase towards tetragonal, out of the MPB region corresponding to a decrease in electromechanical properties.

Utilizing the electrical and mechanical measurements presented, the blocking stress was determined as a function of constant electric field and temperature (Fig. 7). The results from each measurement are presented on the same plot to demonstrate repeatability. It can be clearly observed that at each temperature there was an approximately linear relationship between applied electric field and blocking stress measured as -22.7, -24.2, -27.2, and -28.6 N m⁻¹ V⁻¹ for 25, 50, 100, and 150 °C, respectively. Similar linear blocking force behavior was previously predicted, although not experimentally observed, for bending actuators at room temperature [21]. There have been numerous additional investigations into the observed blocking force-electric field relationship observed in bending actuators [22-25]. In addition,

there is an apparent increase in blocking stress with temperature at each electric field level. This is due to the increase in free strain obtained with temperature as well as changes in the ferroelectric and ferroelastic behavior. At temperatures up to 100 °C there was an increase in free strain, corresponding with the increase in blocking stress. However, at 150 °C there was an observed increase in blocking stress as well as a decrease in free strain. This indicates that the applied electric field at 150°C increased the coercive stress required to ferroelastically switch domains, thereby increasing the stiffness of the sample throughout the tested stress range [18, 26-28]. It is important to note here, however, that the initial Young's moduli at 100 and 150 °C were measured as 36.2 and 36.4 GPa, respectively. The increase in blocking stress at 150 °C was due to the increased effect of the electric field with temperature. With the increase in temperature additional mechanical energy was required to ferroelastically reorient the electrically activated domains, leading to a stiffer system. This also indicates that thermal, in addition to electrical [18], fluctuations will lead to changing actuator stiffness.

The constitutive behavior of the ferroelectric materials used in piezoactuators is nonlinear (Fig. 6). There are two potential significant impacts of nonlinear ferroelasticity on the operation of actuators utilizing ferroelectric materials: (1) the mechanical work done on the load (linear spring) and (2) the spring constant required to impedance match the actuator are different than in the idealized model (Fig. 8). The ratio of ideal to actual mechanical work done on a linear spring is a function of both applied electric field and temperature (Fig. 9a). At low electric field values, or elevated temperatures, the actual work done approaches that of the idealized model (Fig. 9a). In both cases, low electric field and high temperature, this is due to an approximately linear stress-strain behavior in the considered stress region. At low electric field levels (0.5 kV mm^{-1}) the free strain is relatively small, meaning that only the initial portion of the ferroelastic constitutive behavior is important for determination of the blocking stress. At mechanical loads below the coercive stress, approximately -5 to -20 MPa for PZT at room temperature, dependant on dopant content [29-31], approximately linear stress-strain behavior is found. However, at 25 and 50 °C there is a significant observed decrease in the ratio of ideal to actual work at 1 kV mm^{-1} , which is not seen at 100 and 150 °C. This is due to a decrease in coercive field at elevated temperatures [16], which results in an additional electrical stabilization of ferroelectric domains. Previous studies have shown that the coercive stress is more stable with increasing temperature [14], decreasing approximately 20 % from 25 to 200 °C, than the electric poling field [16], which decreases by approximately 51 % from 25 to 200 °C. This indicates that with increasing temperature the influence of the constant electric field increases, while the effect of the stress remains relatively constant. This

necessitates increased mechanical loads to ferroelastically switch an electrically activated domain at higher temperatures, thereby increasing the linearity (stiffness) of the PZT specimens throughout the stress range tested. At each temperature there was a decrease in the ideal to actual mechanical work ratio, although, as seen in Fig. 9a, this ratio is generally larger at higher temperatures. This is due to the increase in stiffness with increasing electric field, with this effect being more dominant at elevated temperatures.

Interestingly, however, despite the increase in the potential maximum work at elevated thermal and electrical loads it was found that the linear spring constant required for impedance matching was approximately equivalent for the ideal and actual material behavior, as shown in Fig. 9b. The approximate maximum variation between the ideal and actual cases was approximately 4%. This effect was not dependent on electric field or temperature, indicating that in terms of determining the ideal impedance matched load the idealized material model is sufficient.

5. Conclusion

The longitudinal strain of a soft PZT commonly used in piezoactuators was measured in response to unipolar electric field and compressive mechanical stress at various temperatures. The electromechanical measurements were performed to determine the blocking stress as a function of electric field and temperature. The significant results are as follows:

- (i) Increasing electric field and temperature both increased the measured blocking force, due to the increase in material stiffness and the increased stabilization of the poled domain state by the applied electric field at elevated temperatures;
- (ii) There was a linear relationship at each temperature tested between the applied electric field and the blocking force;
- (iii) The mechanical work done as computed from the actual nonlinear constitutive behavior exceeded the work computed from the ideal linear spring model used commonly by manufacturers by up to 10 %, depending on temperature and applied electric field;
- (iv) The linear spring constant required to impedance match the PZT material was reasonably determined from the simple linear force-displacement relationship commonly used by manufacturers.

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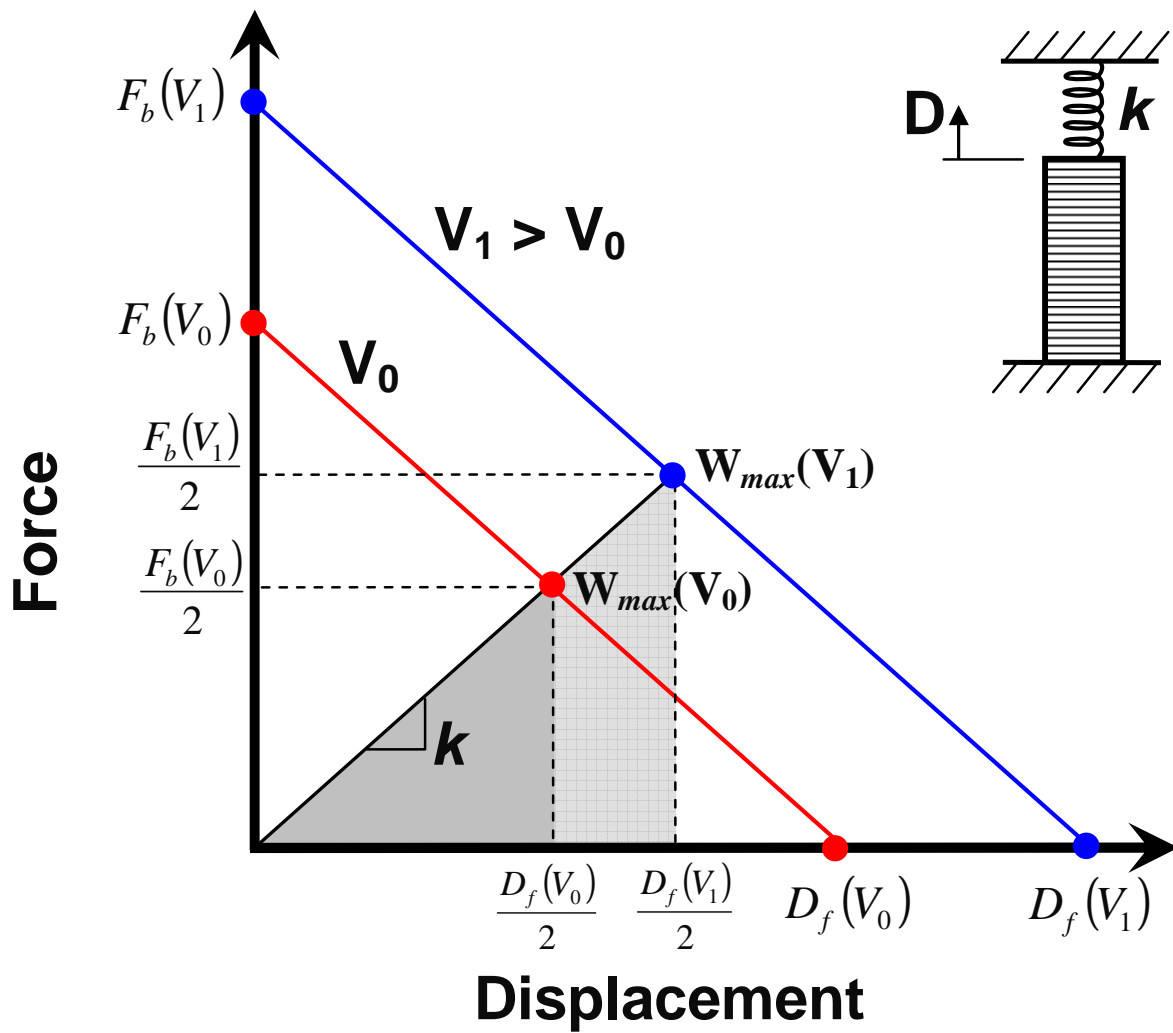


Figure 1. Typical force-displacement curve provided by most piezoactuator manufacturers. Please note that the force is in the compressive direction, whereas it is shown in the figure in the opposite sense.

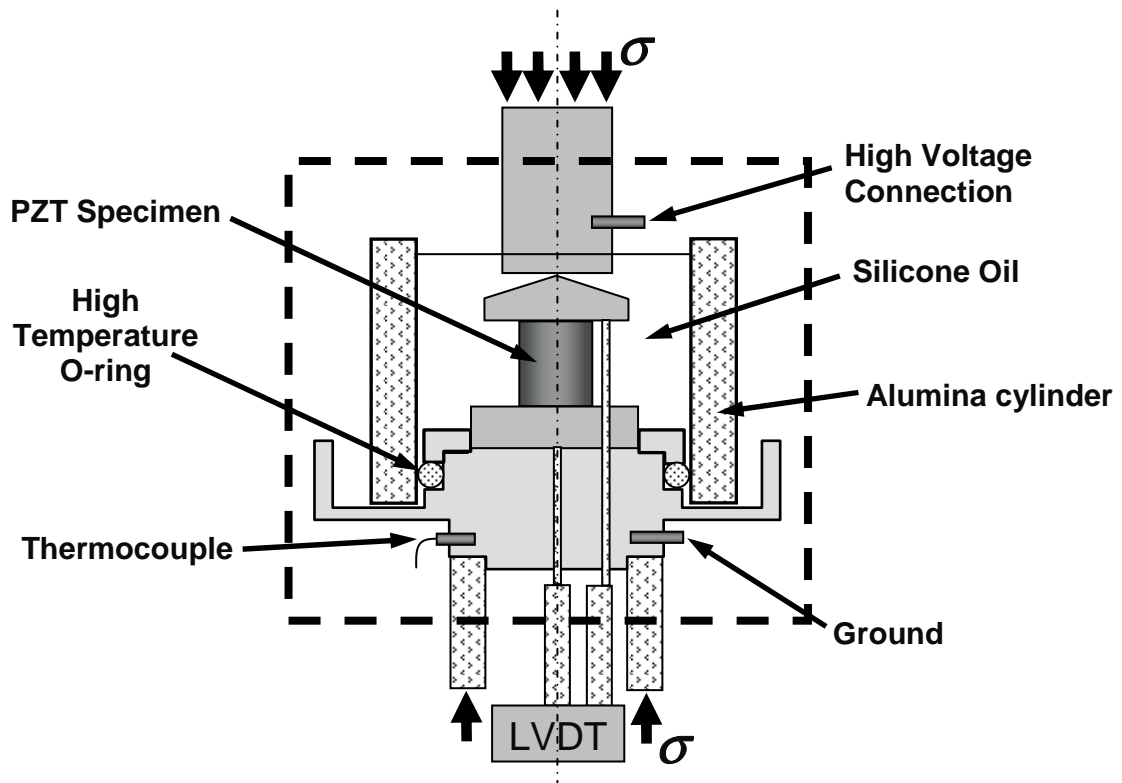


Figure 2. Experimental arrangement for measuring longitudinal strain in response to electric field and stress at elevated temperatures. As an indication of scale the specimen height parallel to the applied load is 6 mm.

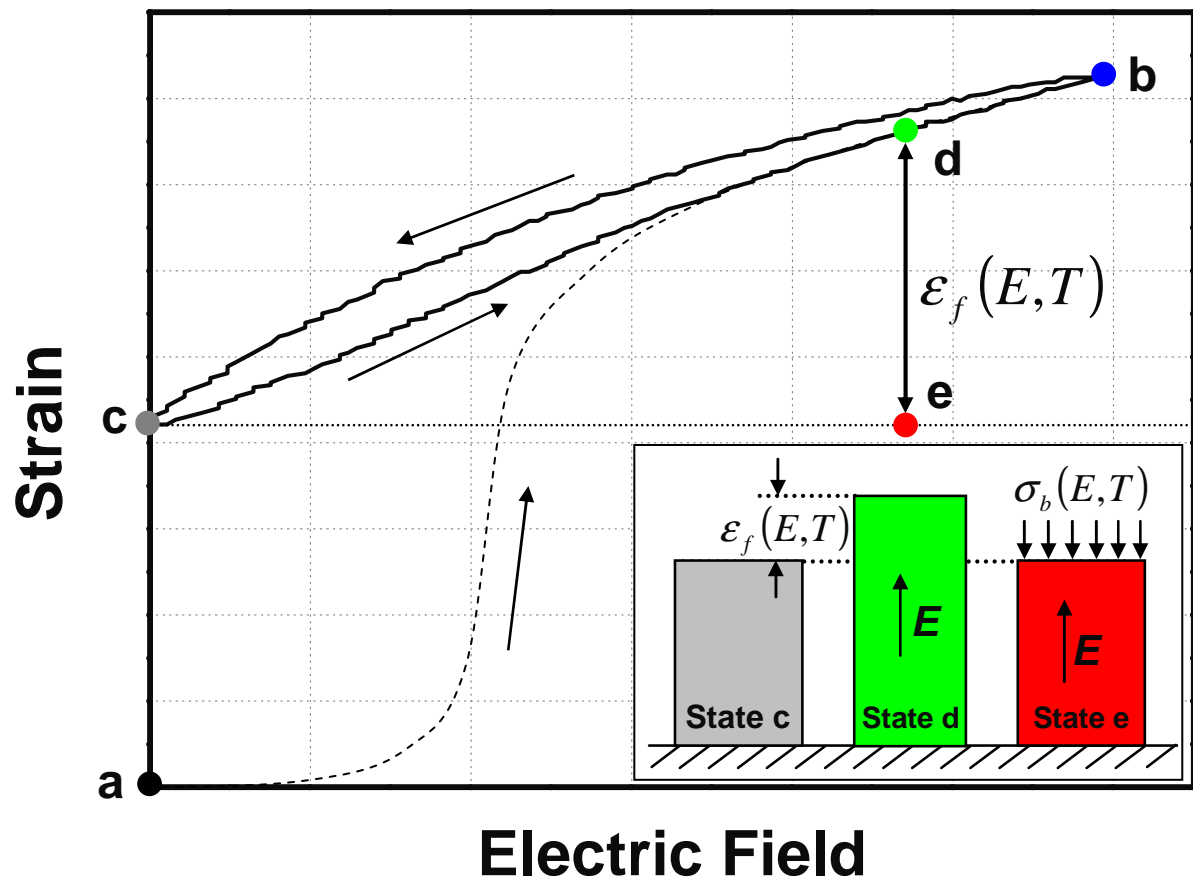


Figure 3. Unipolar electric field-strain hysteresis loop for an initially unpoled sample (Point a). The remnant strain (Point c) was used as the reference in order to determine the free strain of bulk PZT (Point d) as a function of electric field and temperature. The mechanical load required to eliminate the free strain was the blocking stress (Point e). Inset figure displays the three states (c, d, and e) important for determination of blocking stress.

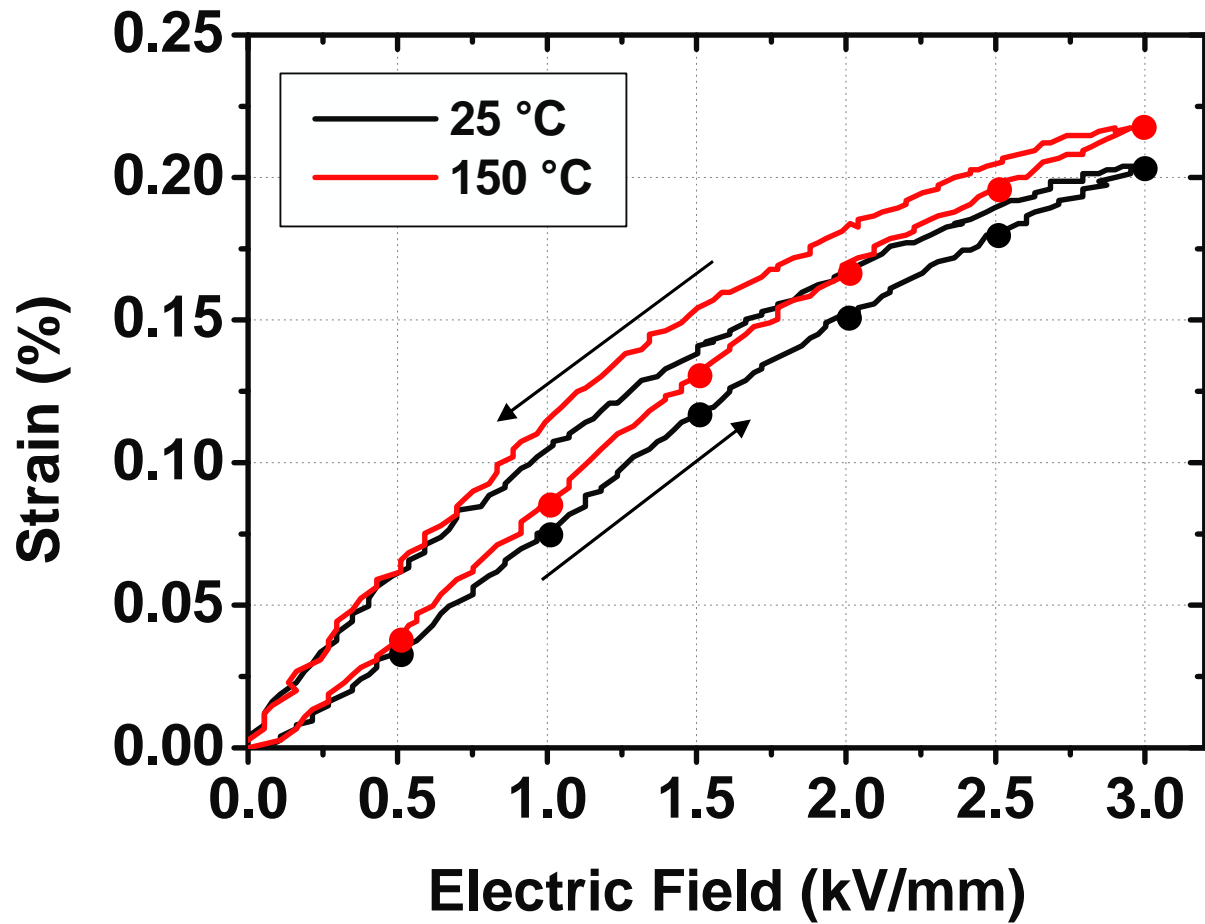


Figure 4. Representative unipolar electric field hysteresis loops for PZT at 25 and 150 °C. The points on the loading path represent the locations where the strain was measured for calibration of the high temperature blocking force measurements. The arrows indicate the direction of loading.

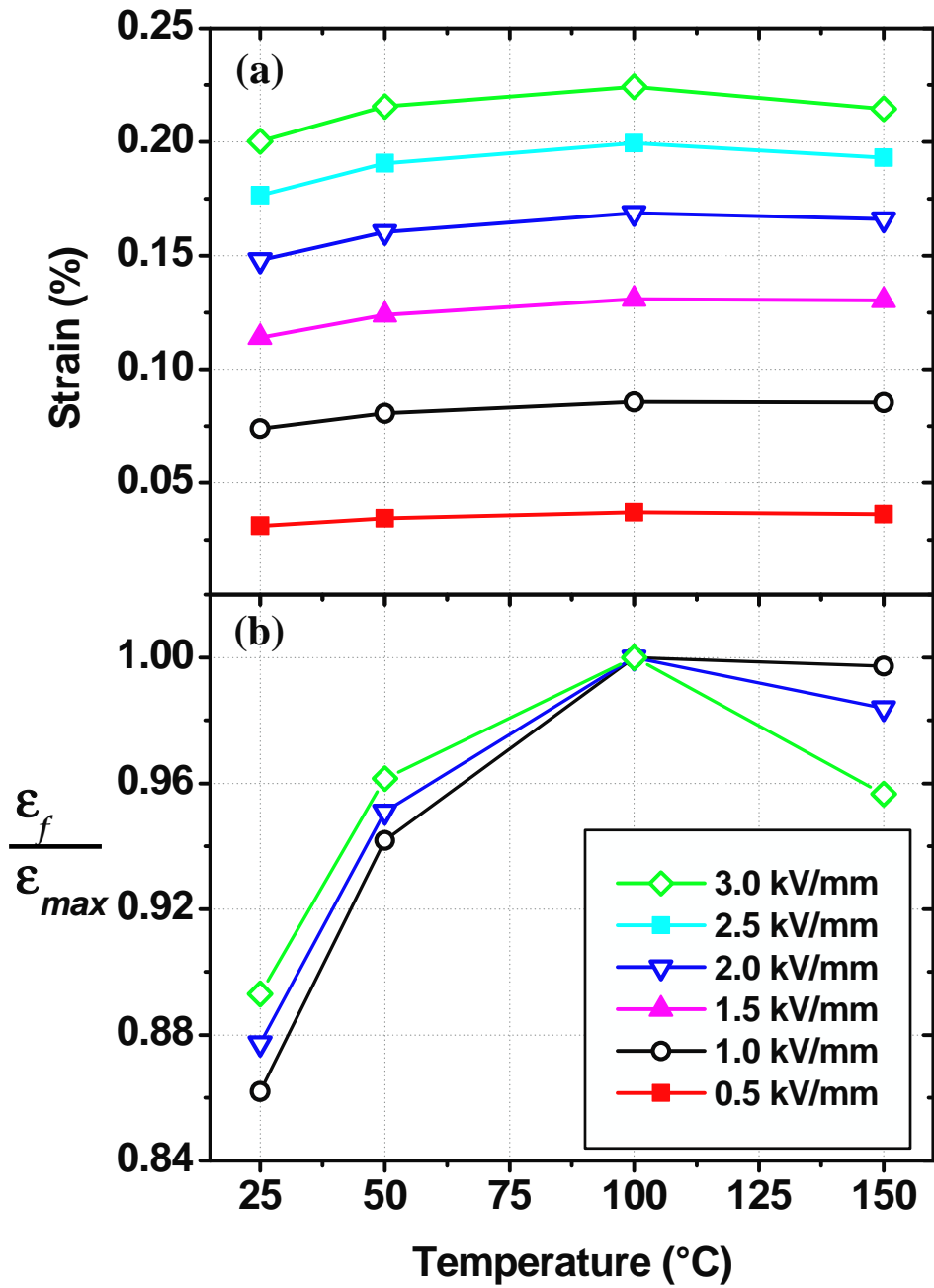


Figure 5. Longitudinal free strain (a) and longitudinal free strain normalized by the maximum strain at the same electric field level (b) measured during quasi-static unipolar electric field loading at various temperatures.

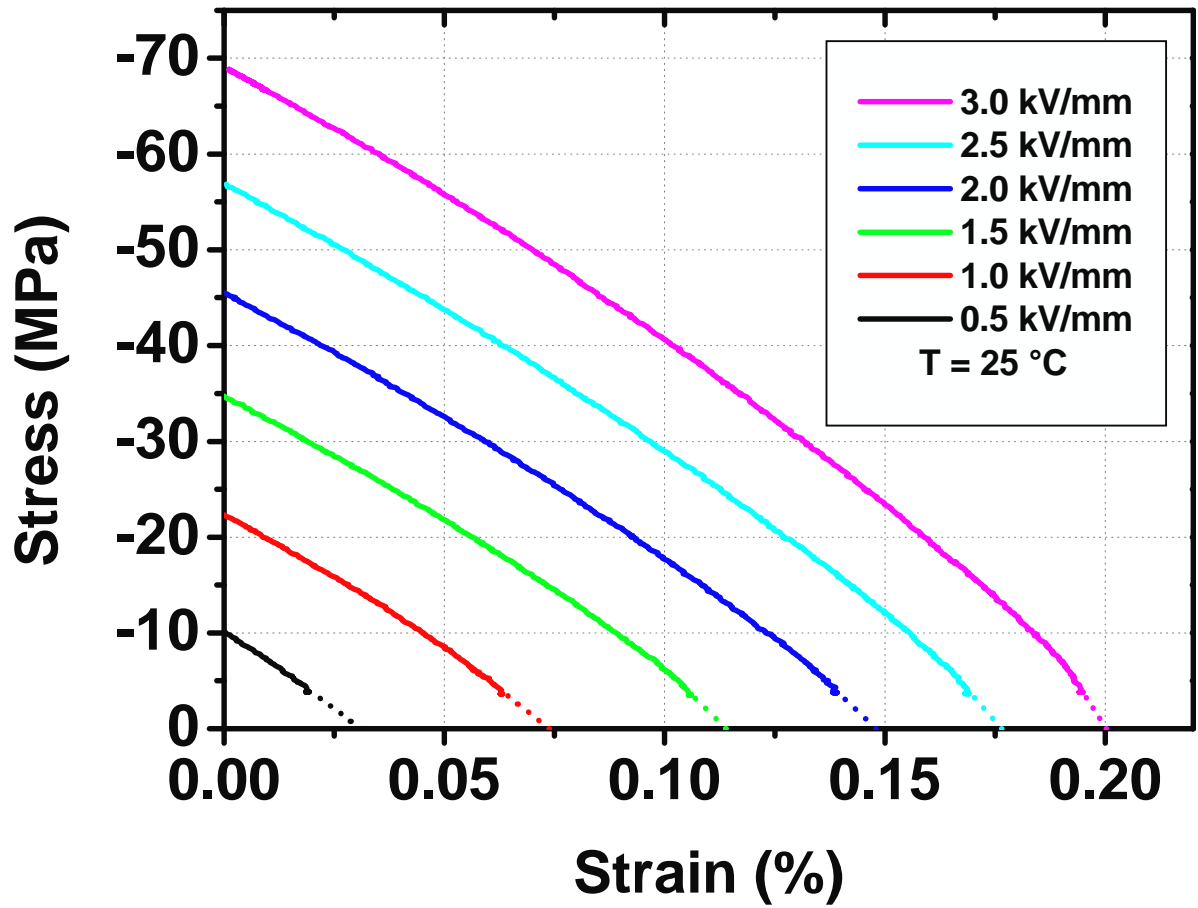


Figure 6. Stress-strain constitutive behavior of soft PZT at 25 °C as a function of constant electric field. Please note that the measured strain does not begin from zero stress because of the required preload. The dotted lines at every electric field level are the measured tangent moduli at the initial stress.

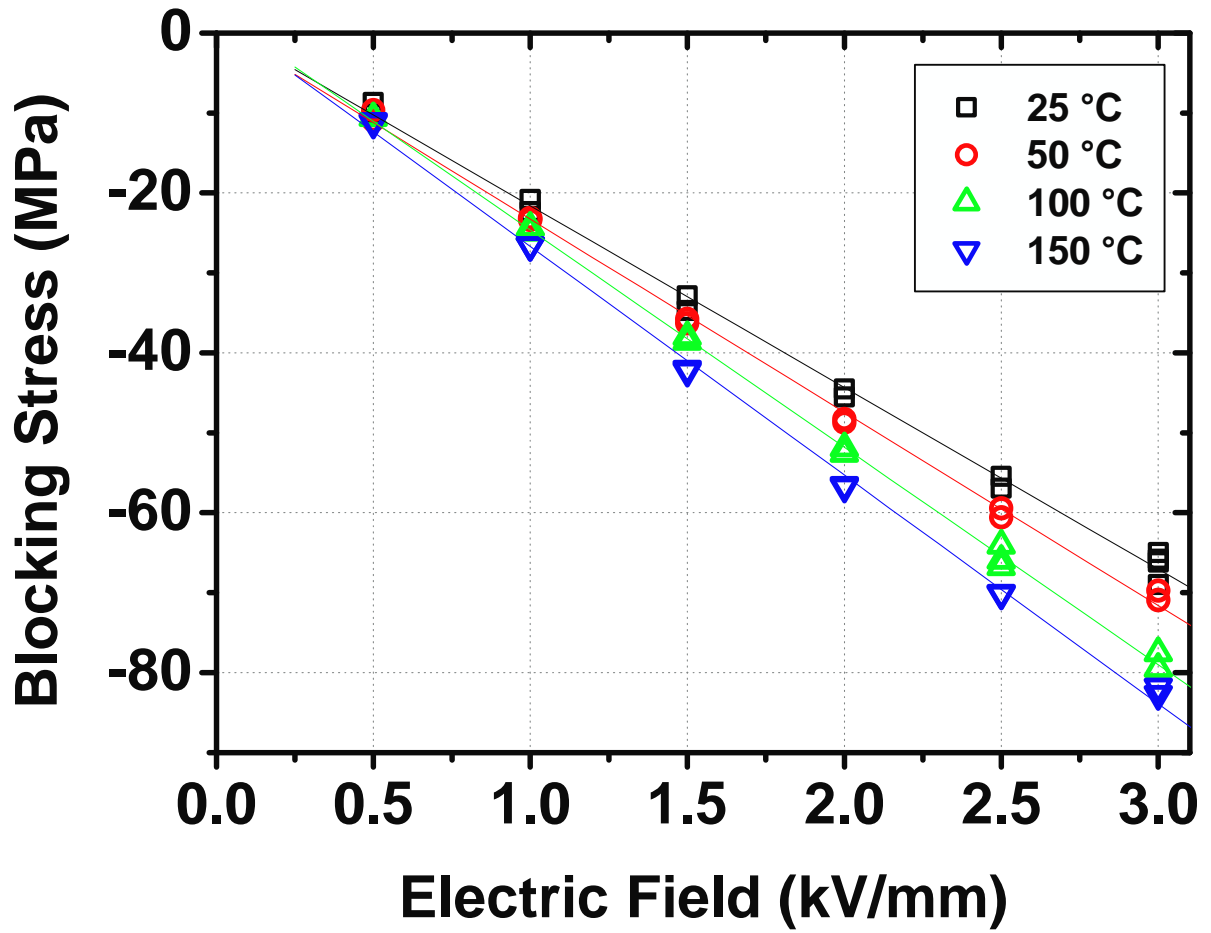


Figure 7. Blocking stress as a function of constant electric field and temperature.

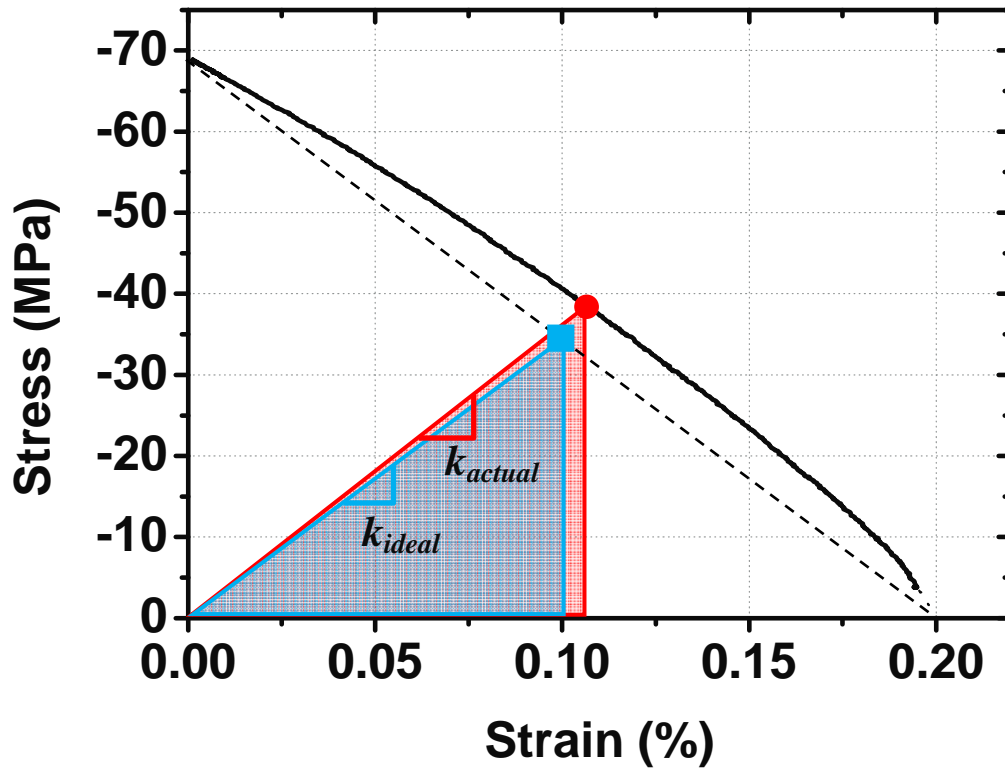


Figure 8. Representative constitutive behavior of PZT at 25 °C (full line) contrasted with the idealized linear material behavior (dashed line). The red (nonlinear stress-strain behavior) and blue (linear stress-strain behavior) triangles represent the maximum work achievable.

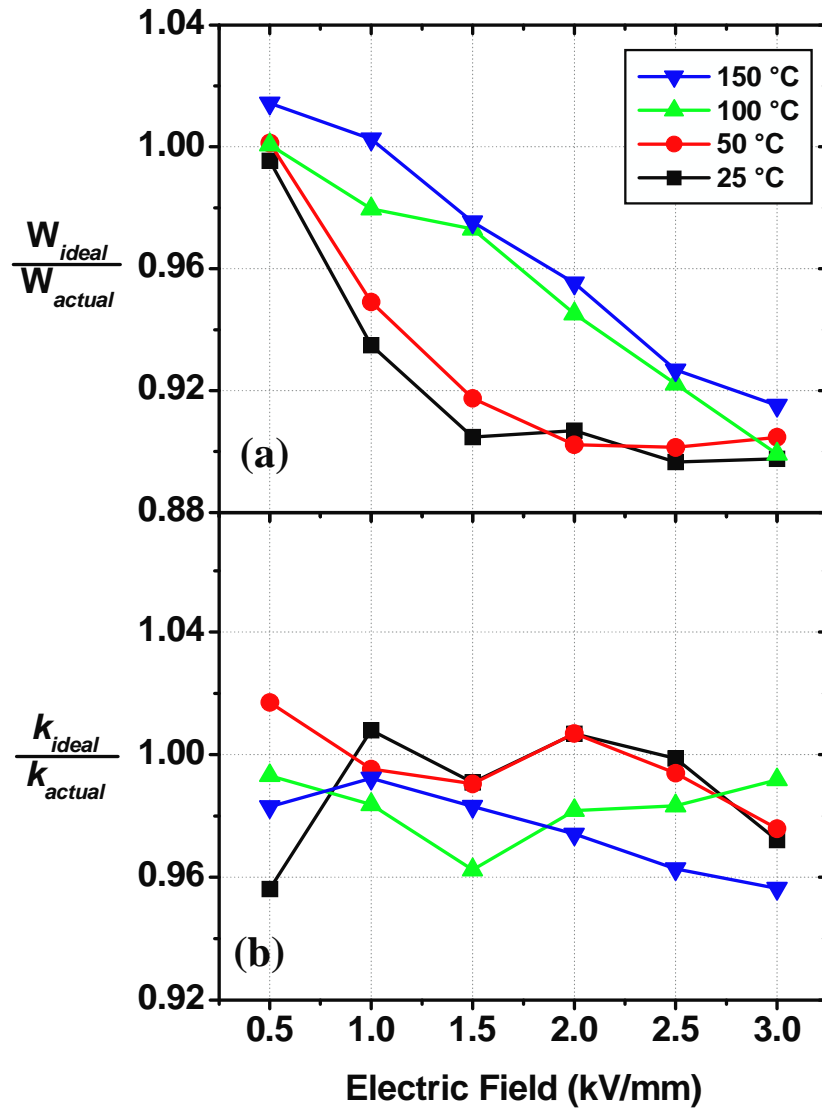


Figure 9. The ratio of the ideal work to the actual work done on a linear spring load (a) and the ratio of the linear spring load required to impedance match the idealized material model to that of the measured nonlinear material behavior (b), both as a function of applied electric field and temperature.