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Modelling of the ferroic material behaviour of piezoelectrics: Characterisation of temperature-sensitive functional properties

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Abstract

Modern multilayer piezoelectric actuators (MPAs) consist of stacks of piezoceramic layers with interdigitated metallic electrodes in between. The combination of thermal, electrical and mechanical loads during service may affect the structural and functional integrity of such electro-mechanical converters. Therefore, a deep knowledge of the coupling phenomena among the field-type quantities such as mechanical stress, electrical field strength and temperature is absolutely necessary. Theoretical simulations are the only way to obtain these (in detail unknown) physical relevant field quantities within MPAs. In this work, the constitutive laws describing the highly non-linear piezoelectric material behaviour are parameterised by experiments, in order to be implemented into FEA tools.

Keywords: Ferroelectrics; Multilayer piezoelectric actuator; PZT; Material properties; Thermo-piezoelectricity

1. Introduction

Lead zirconate titanate (PZT) based piezoceramics are commonly used for technological applications such as low-voltage multilayer piezoelectric actuators (also referred to as MPA) employed in fuel injection systems, for instance.[1] They are designed as laminates of thin PZT-layers separated by interdigitated metallic electrodes. The PZT-material itself is polycrystalline. The crystals exhibit a cubic centro-symmetry at sinter-temperature. When cooling down below the Curie temperature, i.e. less than 350 °C, a phase transition takes place from the paraelectric into the ferroelectric phase. The lattice structure becomes then deformed and asymmetric. As a result, the ferroelectric phase exhibits spontaneous polarisation and strain. Due to the misfit of the distinct variants of the unit cells, the microstructure becomes mechanically stressed and electrically charged and hence twinned in order to reduce mechanical constraints and electric depolarisation fields. In polycrystalline piezoelectrics, the domains of uniform spontaneous polarisation and strain are orientated randomly in the as-sintered state, i.e. no macroscopic piezoelectric behaviour is observable. Because of the ferroic nature of the material, it is possible to force permanent alignment of the different domains using a strong electrical field, the so-called poling process. The so-called "poled state" of the material can be modified by exceeding the electrical, mechanical and thermal limits of the material. In such a case, the ceramic behaves now as a piezoelectric, and the material is remanently polarised and remanently strained (this strain is twice as high as the stroke during application).

In technical applications, piezostacks are assembled by lamination and sintering of metallised piezoceramic layers. These multilayered structures enable the achievement of high electrical field strength causing relatively large strokes with small voltages.[2] Viz., the electrical field strength for MPAs with layer thicknesses of approx. 75 μm and an applied voltage of 150 V is 2 MV/m. The corresponding strokes are in the order of 0.1% (the elongation for a typical piezoelectric actuator with a length of approx. 40 mm is 40 μm). Such piezo-actuators make use of the inverse piezoelectric effect, i.e. an electrical stimulus (voltage) is transformed into a mechanical response (strain). The resulting stroke, which is important for their technical application, is associated with the non-linear ferroic effects such as ferroelectricity and ferroelasticity. Due to a moderate pre-stressing of the MPA, the reservoir of switchable domains may increase because of their alignment perpendicular to an exter-

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nal applied mechanical load, yielding as a result higher strokes and a better actuator-performance. However, although MPAs are mechanically pre-stressed with compressive stresses (−15 to −25 MPa in magnitude) when mounting into the clamp springs, the occurrence of cracks in service has been reported in literature (cracks in ceramics generally propagate only in the presence of tensile stresses).[3] Recent investigations have shown that local tensile stresses may appear in certain critical regions of the MPA (e.g. active–passive-zone, electrode-tips) owing to the coupled thermo-electro-mechanical stresses during fabrication, poling and functioning of the MPA.[4]

During the sintering of MPAs internal tensile stresses may result from the densification process, since the field-free passive-zone is constrained by the electrode layers in the active zone. The internal stresses concentrate around the electrode-tips. Further internal stresses are generated by poling the piezoceramic because the domain-alignment (i.e. the inelastic deformation) in the active zone is constrained near the free electrode edges by the adjacent passive-zone, where only mechanically induced strains occur. In fact, it has been reported in the literature that cracks along the electrodes may appear during poling due to the strain misfit of these adjacent regions.[7]

During the actuating process the behaviour of multilayer piezoelectric actuators is dominated by the non-linear ferroelectric effect (the MPA is subjected to a strong electrical field strength as the driving force for domain-processes). When no external electrical field is applied, their behaviour is dominated by ferroelastic effects, cf. Fig. 1.

In service, low electrical field strengths induce a nearly linear piezoelectric strain effect (low-signal range or in the dynamic regime), whereas in the large-signal (quasi-static) regime a non-linear strain response is caused by the realignment of the polar axes of the crystals (domain switching effects). The alignment of the domains causes an inelastic deformation,[5,6] which – with respect to the occurrence of internal stresses – can be treated analogously to metal plasticity. Like in the case of fatigue in metals, a cyclic inelastic deformation and Joule heating occur. These effects lead also to periodic tensile stresses concentrated in the vicinity of electrode-tips. Hence, a cyclic loading induces fatigue damage (the growth of cracks) in the piezoceramic material.

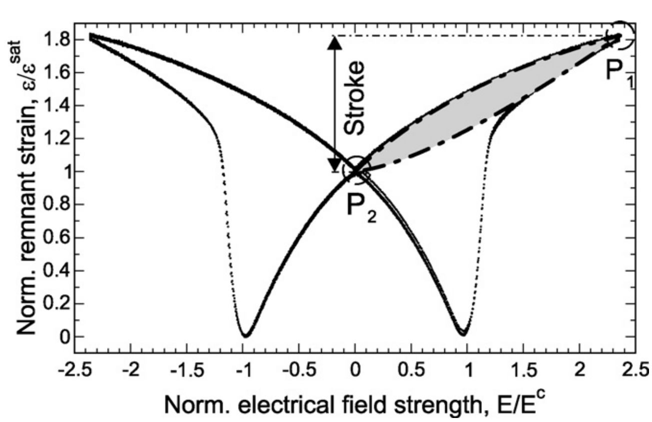

Fig. 1. MPAs make use of the inverse piezoelectric effect. A strong electrical field causes a remanent strain. During acting as actuator (unipolar cycling; grey coloured hysteretic loop) their behaviour is dominated by the non-linear ferroelectric effect, P_1. When no external electrical field is applied, the ferroelastic effect, P_2, dominates all other effects. Viz., the stroke is strongly influenced by these non-linearities.

Based on the complex loading scenario in an MPA, a detailed knowledge of the coupled phenomena among the field-type quantities mechanical stress, electrical field strength and temperature is absolutely necessary. A theoretical simulation is the only way to obtain all relevant physical field-quantities within MPA-devices. Hence, for structural–mechanical FE-analyses of the highly non-linear behaviour of piezoelectrics, ferroelectrics fundamental modelling parameters have to be evaluated. In this work, the implementation of the constitutive material laws[6,8] into FEA-tools has been assessed by experimental determination of the material properties on a homogenous commercial PTZ bulk ceramic under distinct thermo-electro-mechanical loading conditions.

2. Nomenclature

In order to ease the readability of this paper, the nomenclature list, confer Table 1, maps the commonly used abbreviations for the relevant physical quantities to avoid any confusion.

3. Theoretical approach: the model

The basic assumption in order to model the non-linear piezoelectricity is the additive decomposition of the dielectric displacement, $\mathbf{D}$, and total strain, $\boldsymbol{\varepsilon}$ into reversible and irreversible (or remanent) contributions[8,9]:

$$\begin{aligned} \boldsymbol{\varepsilon} &= \boldsymbol{\varepsilon}^{rev} + \boldsymbol{\varepsilon}^{i} \\ \mathbf{D} &= \mathbf{D}^{rev} + \mathbf{P}^{i} \end{aligned} \quad (1a)$$

where the $\mathbf{P}^i$ is the remanent dielectric displacement. The reversible part can be represented by the linear piezoelectricity.

Table 1
Nomenclature list.

Letter	Physical meaning
$\mathbf{C}$	4th-rank elasticity tensor
c^E	Dielectric hardening function due to external applied electrical field
$\mathbf{D}$	Total dielectric displacement
$\mathbf{d}$	3rd-rank piezo-modulus (piezoelectric voltage constants)
E^C, $\mathbf{E}$	(Coercive) electrical field strength
f^E	Onset for domain switching due to external applied electrical field
h^E	Offset constraint; fully poled material state
$\mathbf{P}$	Total polarisation
P^{sat}	Maximum achievable remanent polarisation for electrical field strengths up to 2 MV/m
γ	Curie constant
$\boldsymbol{\varepsilon}$	Total strain
ε^{sat}	Maximum achievable remanent strain due to poling the piezoelectric
δ	Kronecker-delta
$\boldsymbol{\kappa}$	2nd-rank permittivity-tensor
$\boldsymbol{\sigma}$	Total stress
σ^C	Coercive stress

Thus, Eq. (1a) can be rewritten as:

$$\begin{aligned} \boldsymbol{\varepsilon} &= \mathbf{C}\boldsymbol{\sigma} + \mathbf{d}\mathbf{E} + \boldsymbol{\varepsilon}^i \\ \mathbf{D} &= \mathbf{d}\boldsymbol{\sigma} + \boldsymbol{\kappa}\mathbf{E} + \mathbf{P}^i \end{aligned} \tag{1b}$$

with $\mathbf{C}$ as 4th-rank elasticity tensor, $\mathbf{d}$ as 3rd-rank piezo-modulus, $\boldsymbol{\kappa}$ as 2nd-rank permittivity-tensor, and $\boldsymbol{\sigma}$ and $\mathbf{E}$ as the mechanical stress and electrical field vector, respectively. Note that the piezoelectric voltage coefficients and the permittivity are strongly dependent on the poling state of the piezoelectric material. Their magnitudes are linearly rescaled from the values in the isotropic material state (the piezoceramic does not behave as a piezoelectric due to no macroscopic polarisation) up to the measured values of the fully poled anisotropic state.[8]

The linear thermo-piezoelectricity is reversible in the sense that it vanishes in the absence of external loading (e.g. by mechanical stresses or electric field strengths). The irreversible quantities, as internal model variables, represent the remanent states of the material resulting from an average of the microscopic domain configurations onto the macroscopic level. The remanent parts are further decomposed into a part caused by externally applied electrical field strength and into mechanical components (representing ferroelasticity and the corresponding de-poling) exceeding the material's coercive threshold values (E^C or σ^C) for the initiation of domain switching effects. If the external mechanical loads, $\boldsymbol{\sigma}^{apl}$, do not exceed the coercive stress, σ^C, the mechanical components are zero per definition. This is the case for moderately pre-stressed piezostacks, where $\boldsymbol{\sigma}^{apl} \approx \sigma^C/3$, in the first approach.

The constitutive law utilised in this work is based on concepts of plasticity; sc. a switching criterion, an associated flow rule and purely kinematic hardening to describe the cyclic response. Hence, the evolution equations are defined on the basis of the following switching criteria,[8,9]

$$\begin{aligned} f^E &= \left\|\mathbf{E} - c^E\mathbf{P}^E\right\| - E^C \\ h^E &= \left\|\mathbf{P}^E\right\| - P^{sat} \end{aligned} . \tag{2}$$

The former equation, f^E, indicates the onset of the domain switching due to electrical loading and the latter, h^E, represents the fully switched ferroelectric domain states, respectively, i.e. the material hardens because, as the process of domain switching proceeds, internal constraints in the polycrystalline compound increase and the reservoir of switchable domains is reduced. Hence, the "hardening", c^E, depends on the internal state of the material due to the remanent polarisation. Thus, the governing evolution law for the internal variable can be written as

$$\dot{\mathbf{P}}^E = Tr_1(f^E)Tr_2(h^E)\frac{\dot{\mathbf{E}}}{c^E(\mathbf{P}^E)}, \tag{3}$$

with $Tr_{1,2}$ as trigger-criterion for the onset and the saturation of the domain switching process. The corresponding piezo-strain is caused by the remanent polarisation, hence,

$$\varepsilon_{ij}^{\mathrm{P}} = \frac{3}{2}\varepsilon^{sat}\frac{\left\|\mathbf{P}_k^E\right\|}{P^{sat}}\left(\mathbf{e}_i^{\mathrm{P}}\mathbf{e}_j^{\mathrm{P}} - \frac{1}{3}\delta_{ij}\right). \tag{4}$$

The superscript *sat* represents the maximum remanent internal state and $\mathbf{e}_i^{\mathrm{P}}$ the unit polarisation-vector.

In summary, the basic modelling parameters that have to be measured are: remanent polarisation and strain, coercive electrical field strength, hardening function, relative permittivity, piezoelectrical voltage coefficients and the effective elastic modulus (as function of temperature).

4. Experimental: measurements

4.1. Material of study

The following measurements enable the derivation of crucial material parameters to compute the ferroelectric hysteresis-curves in an appropriate way by only taking into account kinematic hardening. Therefore a commercially available pure rare earth (RE-) doped PZT-ceramic – as the basis material for the multilayered piezoceramic stacks – was tested. This material is a RE_2O_3–$PbTiO_3$–$PbZrO_3$ ternary phase system incorporating approx. 2–3% of the rare earth oxide in the vicinity of the morphotropic phase boundary (MPB) of PZT in the tetragonal range. The rare earth in the composition acts as a donor to make the material "soft."

The ceramic sheets were stacked without electrodes in between with a resulting dimension of 3.5 mm × 3.5 mm × 40 mm. The bulk ceramic was sintered at approx. 1100 °C in a lead-enriched atmosphere (with the same sintering-parameters as for the multilayered piezostacks). Thereafter the sintered bodies were cut into distinct specimen geometries, according to the piezoelectricity standards.[10] External Cr–Ag-metallic layers were sputtered onto the specimen by a PVD-process acting as electrodes. To eliminate mechanically induced domain orientations, the samples were annealed at 400 °C for 1 h after mechanical preparation.

4.2. (Ferro-)electric characterisation

The comprehensive electrical and electro-mechanical characterisation of piezoelectric bulk ceramic samples was done with a Piezoelectric Evaluation System (aixPES) provided by aixACCT Systems GmbH, Germany. The specimens were (three times) bipolar poled with electrical field strengths in the order of approx. 2 MV/m from ambient temperature up to 150 °C (i.e. the estimated operating temperature). This enables the identification of the modelling parameters such as the coercive electrical field strength, E^C, the irreversible quantities remanent polarisation, $\mathbf{P}^i$, and strain, $\boldsymbol{\varepsilon}^i$, as internal variables dependent on temperature,[5,6] and the kinematic hardening condition.

Large-signal material features were evaluated over a wide temperature range. The sample's current response was measured by applying an electrical voltage excitation signal using the flexible and precise virtual ground method. The displacement was simultaneously measured with a laser interferometer system. The influence of the temperature on the electro-mechanical coupling in the small signal range was assessed by means of a temperature chamber (Carbolite GmbH, Germany) coupled to an impedance analyser (Agilent, 4294A). The temperature tests were performed following a ramp with a heating rate of 1.5 °C/min ranging from room temperature (RT) up to 400 °C.

The material's parameters were determined using weak-field (<0.01 MV/m) techniques such as the resonance antiresonance method (IEEE standard).[10]

4.3. (Ferro-)elastic characterisation

The mechanical characterisation was done with a universal testing machine (UTM) MIDI 10-5 7 × 13″ from Messphysik, Austria. An adaptation of this UTM enabled the measurement of both mechanical quantities such as mechanical stresses and the corresponding strains, as well as electrical quantities such as charges (or polarisation) and the applied electrical voltage (electrical field strength) within the MPAs. For bulk ceramics only mechanical measurements were possible because of a limitation of the voltage source supply of 1.5 kV.

For ferroelastic studies uniaxial compression tests were performed on initially non-poled piezoceramic bar-shaped specimens (3.5 mm × 5 mm × 25 mm) without electrodes.[6] The aspect ratio of nearly 5:1 ensures that a significant central region of the sample experiences uniform uniaxial stress and strain states. The external load-induced deformations (longitudinal strain) were monitored using a pair of strain gauges mounted on opposite sides of the specimen. The measurements were repeated three times to ensure reliability of the experimental results.

In addition, the Young's modulus (in this case called "tangent modulus") was evaluated from the compression stress–strain curves. Since the piezoelectric effect is a coupling phenomenon between the electrical field strength, mechanical stresses and temperature (the latter associated with the distinct phase state), the elastic modulus and corresponding damping effects were characterised in the relevant temperature operative range of application, i.e. from −50 °C up to 150 °C. The temperature tests were performed following a ramp with a heating rate of 1 °C/min and a dwell of 10 min at the target temperature.

5. Results and discussion

5.1. (Ferro-)electric response

Fig. 2 shows the dielectric displacement and the corresponding strain vs. the electrical field strength of the PZT-material measured at different temperatures. It can be inferred that by exceeding the ferroelectric threshold value for domain switching, E^C, the PZT-material becomes remanently polarised and strained. The material hardens, i.e. the reservoir of "switch-able" domains diminishes. After reaching a saturation-plateau (at approx. 2 MV/m) nearly all domains align parallel to the external applied electrical field strength. At heightened temperatures the coercive and remanent quantities diminish, as can be inferred in Fig. 2 (cf. the corresponding shift inwards).

The remanent quantities such as remanent polarisation and remanent strain can be fitted by a Curie Weiss-law as function of temperature:

$$\frac{P^i}{P^i_{RT}}\left(\frac{\varepsilon^i}{\varepsilon^i_{RT}}\right) = \gamma\sqrt{T_C - T}, \quad (5)$$

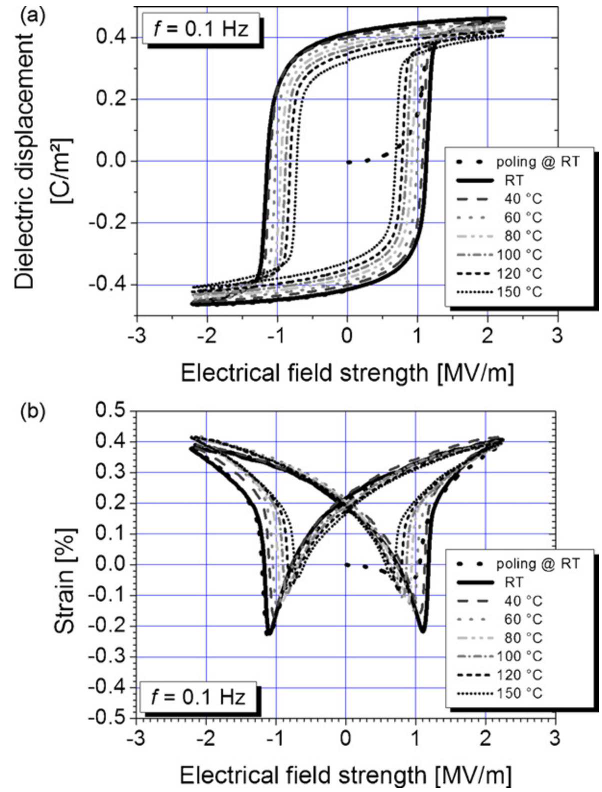

Fig. 2. By exceeding the ferroelectric threshold value for domain switching, E^C, the PZT-material becomes remanently polarised and strained. The material hardens, i.e. the reservoir of "switch-able" domains diminishes. After reaching a saturation-plateau (at approx. 2 MV/m) nearly all domains align parallel to the external applied electrical field strength. At heightened temperatures the coercive and remanent quantities diminish (cf. the corresponding shift inwards) but the values for the permittivity and the piezoelectric voltage coefficient (slope becomes steeper) increase. (a) The dielectric and (b) the corresponding "butterfly"-loops.

with γ (≈5.75 × 10^{-2}) as Curie constant and T_C as Curie temperature (i.e. 350 °C). The coercive electrical field strength behaves nearly linear with the temperature, T.

Furthermore, the piezoelectric voltage coefficients and the permittivity are strongly dependent on temperature and electrical field strength as well as frequency due to time-dependence of domain switching, cf. Fig. 3. In the dynamic regime, domain switching can be neglected, whereas in the quasi-static range it plays an important role. Experimental findings showed that these quantities increase significantly with the temperature, especially when approaching T_C. Nevertheless, for the operative range, the values of permittivity and piezoelectric voltage constants can be easily fitted by a quadratic polynomial.

5.2. (Ferro-)elastic response

Fig. 4 shows the strain variation of the material under compressive stress. Considering the tangent modulus (Young's modulus) through the entire loading range, a strong change in the elastic response of the material under uniaxial compressive stress loading can be appreciated. Young's modulus was found

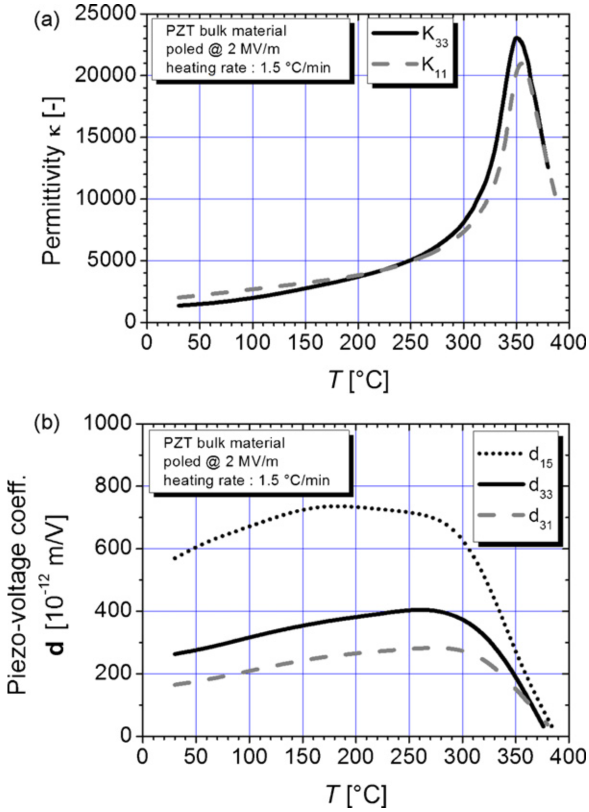

Fig. 3. Dependence of the permittivity (a) and the piezoelectric voltage coefficients (b) on temperature, dynamically measured (kHz-range). At evaluated temperatures these quantities increase significantly up to close to Curie temperature.

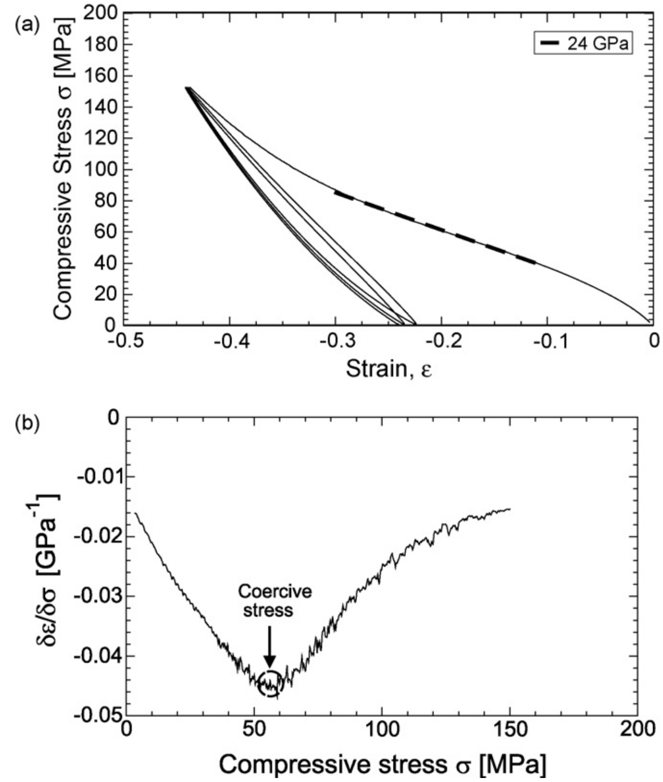

Fig. 4. Uniaxial compression tests were performed on piezoceramic bodies (3.5 mm × 5 mm × 25 mm) without electrodes in between. The stress–strain loops indicate the strongly non-linear ferroelastic behaviour (a). At the coercive stress a maximum number of domains align perpendicular to the external applied mechanical load; the value of the tangent modulus reaches a minimum at this point (b).

to increase from 70 GPa up to 130–150 GPa with increasing stresses in the depolarised material state. The resulting coercive stress (defined as the stress where a maximum number of domains align perpendicular to the external applied load), $\sigma^C = -57$ MPa, is approx. three times the typical pre-stress of a multilayer piezoelectric actuator (approx. −20 MPa). A minimum in the Young's modulus is found at the coercive stress. Here the tangent modulus is only 24 GPa in magnitude because most of the domains are prone to switch perpendicular to the external applied mechanical load. Table 2 reviews basic modelling parameters evaluated through the experiments at room temperature.

Fig. 5 shows the stability of the effective elastic modulus obtained by using the resonant beam technique.[11] It can be seen that it remains nearly constant up to the Curie temperature, i.e. approx. 350 °C, where the phase transition takes place. At higher temperatures, however, the elastic modulus doubles compared with the at room temperature value (RT). Regarding the damping, it can be measured below Curie temperature over the whole range of temperature of study. It can be speculated that the thermal activation during heating eases domain switching, leading to damping effects. On the other hand, when cooling down from the Curie temperature, the microstructure may become twinned

Table 2
Basic parameters at room temperature to model the non-linear piezoelectricity.

Coercive electrical field strength	1.1 MV/m
Maximum achievable remanent polarisation	0.4 C/m^2
Maximum achievable remanent strain	0.2%
Piezoelectric modulus (**d**$_{33}$); large-signal	850 pm/V
Relative permittivity (K$_{33}$); large-signal, poled	2440
Coercive stress	57 MPa

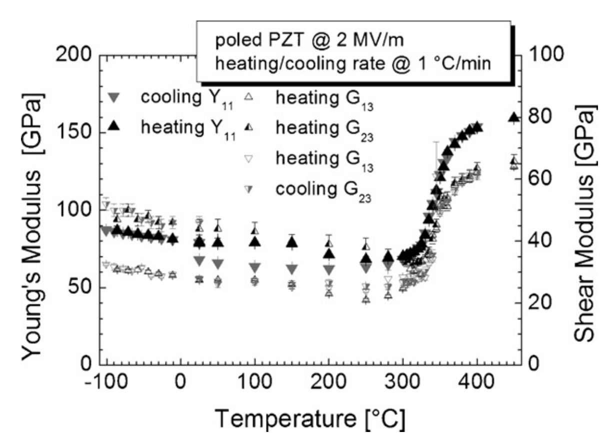

Fig. 5. The effective elastic modulus is nearly constant up to the Curie temperature, i.e. approx. 350 °C, where the phase transition takes place. At higher temperatures, the effective elastic modulus becomes double the value at room temperature (RT). The symbols ▲, and ▼ represent the heating and cooling parts of the cycle, respectively.

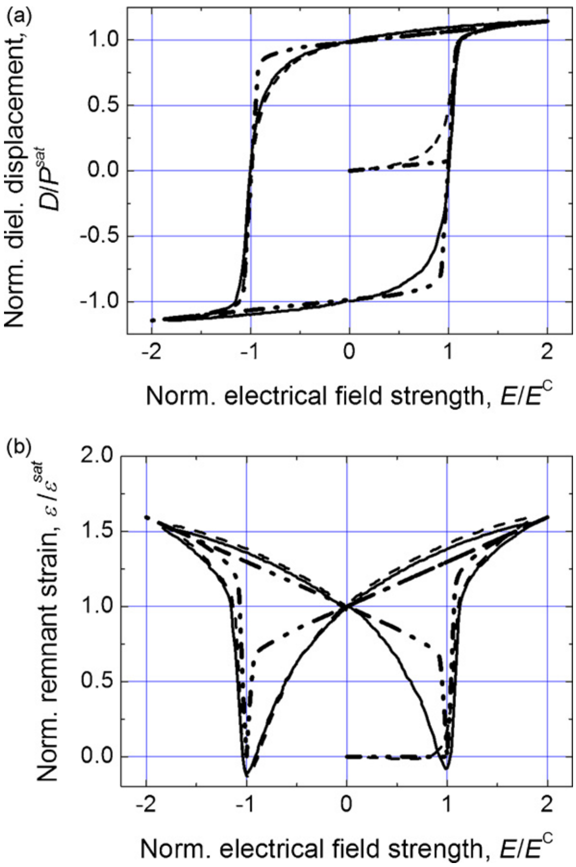

Fig. 6. Comparison of the computed results (dash dot dot) with the experiments (dashed; solid line). The dashed line indicates the poling loop of a virgin material at ambient temperature and, the solid line the 2nd bipolar loop; (a) the dielectric hysteresis-loop and (b) the "butterfly"-curve.

in order to reduce mechanical constraints and electric depolarisation fields by domain wall movements, causing as a result mechanical losses.

5.3. Comparison of computed results and experiments

Fig. 6 shows the dielectric hysteresis-loop and the "butterfly"-curve of the material of study in a combined plot, presenting the computed results (dash dot dot) and the experiments (dashed; solid line). The dashed line indicates the poling loop of a "virginal" PZT-material at ambient temperature and the solid line corresponds to the 2nd bipolar loop. When the external electrical field strength overcomes the ferroelectric threshold value for the initiation of domain switching, i.e. approx. 1.1 MV/m, the PZT-material becomes remanently polarised and strained. The piezoelectric hardens because the reservoir of "switchable" domains diminishes. After reaching a maximum remanent polarisation, i.e. approx. 0.4 C/m^2, the alignment of the domains is equivalent to the field vector, and the material behaves nearly linear elastic.

A comparison of the modelled loops with the experiments (done on pre-stressed PZT-specimens) shows consistency in the basic model assumptions. Ongoing FE-simulations based on these constitutive laws will enable the analysis of critical realistic loading scenarios.

6. Conclusions

In this study basic modelling parameters have been determined enabling the parameterisation of constitutive laws reported in literature.[8] A comparison of the modelled loops with the experiments shows consistency in the basic model assumptions for ambient temperatures. In future the strong temperature dependence of the modelling parameters will be also taken into account. For moderately pre-stressed piezostacks this model is satisfactory for describing the poling behaviour. In future investigations the ferroelastic, thermal coupling effects and a combination of both the isotropic and kinematic hardening will be considered and implemented into a FE-code.

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