Measurement System for Piezoelectric Resonance Impedance Spectroscopy Under Combined AC and High-Voltage DC Loading

Tomaž Kos, Mihail Slabki, Janko Petrovčič, Damir Vrančić, Gregor Dolanc, and Jurij Koruza, Senior Member, IEEE

Abstract—Piezoelectric resonance impedance spectroscopy is a standardized measurement technique for determining the electromechanical, elastic, and dielectric parameters of piezoceramics. However, commercial measurement setups are designed for small-signal measurements and encounter difficulties when constant driving voltages/currents are required at resonances. Combined AC and DC loading. The latter is particularly important to evaluate the DC bias-hardening effect of piezoceramics. Here, we propose a novel measurement system for piezoelectric resonance impedance spectroscopy under combined AC and high-voltage DC loading that complies with established standards. The system is based on two separate output amplifier stages and includes voltage/current probes, a laser vibrometer, custom protection components, and control software with optimization algorithm. In its current form, the measurement setup allows the application of AC frequencies up to 500 kHz and DC signals up to ±10 kV on samples with impedance between 10⁻¹ and 10⁸ Ω. The operation of the proposed setup was benchmarked against commercial impedance analyzers in the small-signal range and reference equivalent circuits. Test measurements under combined AC and DC loading were performed on a soft Pb(Zr,Ti)O₃ piezoceramic. The results revealed that a DC bias voltage applied along the polarization direction ferroelectrically hardens the material, while the material softens and eventually depolarizes when the DC bias voltage is applied in the opposite direction. The results confirm the suitability of the designed measurement system and open new exciting possibilities for tuning the piezoelectric properties by DC bias fields.

Index Terms—Ferroelectrics, impedance spectroscopy, piezoceramics, resonance measurement.

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1. INTRODUCTION

Piezoceramics are widely used in sensors, actuators, and transducers, enabling advanced technologies in industrial automation, consumer electronics, smart vehicles, micro robotics, and medicine [1]. The highest piezoelectric properties are obtained in ferroelectric materials, where the electromechanical conversion mainly originates from the crystal lattice and ferroelectric domains [2], [3]. While the latter are important and often dominant contribution to strain, they are also largely responsible for hysteretic behavior and losses [4], [5]. These are detrimental to the use of piezoelectrics under near-resonance conditions, e.g., in ultrasonic motors and transducers. Ferroelectric hardening refers to the process of limiting domain wall motion by point defects [6]–[8], secondary phases [9], or precipitates [10]. These concepts enable the use of piezoceramics under resonant conditions that allow the generation of large oscillatory strain amplitudes and high vibration velocities at relatively low applied electric fields. The most widely used approach is ferroelectric hardening by point defects introduced into the perovskite lattice by acceptor doping. However, this mechanism is also susceptible to degradation at elevated temperatures and field amplitudes, mainly related to the mobility of oxygen vacancies [11], [12] and the large contribution of domain walls in lead-based piezoceramics [5], [13].

An alternative approach for ferroelectric hardening is based on DC bias fields. Nonlinear polarization and strain response to external electrical or mechanical loads are among the prominent features of ferroelectric materials. Therefore, it is obvious that ferroelectric properties can be modified by superimposing static mechanical stress or DC bias voltage on the driving AC stress/voltage signal. Mechanical preloading is a well-known approach for actuators [14]–[16] and ultrasonic transducers [17]. The impact of external perturbation by DC bias on electromechanical properties has been studied under off-resonance [18]–[20] and resonance conditions [21]. It was demonstrated that ferroelectrics undergo pronounced hardening when a DC bias voltage is applied along the polarization direction and softening when the field is applied in the opposite direction. The observed ferroelectric hardening is related to the immobilization of the domain walls by the external field and the alignment of the polarization vector along the field direction. It should be noted that from the application point of view, the DC bias voltages are arguably easier to implement...
for tuning material properties compared to mechanical stress, since they can be superimposed on the driving AC voltages and used in positive or negative polarity. However, mechanical stress is usually implemented only as compressive loading, since tensile stress in the megapascal range is difficult to apply and leads to earlier fracture of piezoceramics due to low fracture toughness [22].

Although DC hardening is an interesting alternative to conventional hardening approaches, the effects of DC fields on coupling coefficients and mechanical quality factors are poorly understood and very few studies exist. One of the main reasons is the difficulty of combining large DC and AC signals under resonance conditions. Indeed, commercial measurement systems do not provide the required combination of wide frequency range, high DC voltage bias, and drive current capability. Most devices use the same output amplifier stage for both voltage signals (DC and AC); therefore, devices with high DC voltage capability have high output impedance, low current capability, and narrow frequency range. On the other hand, wide bandwidth devices have low output impedance but are limited by low DC voltage capability. In addition, these devices are not equipped with a simultaneous measurement of the mechanical vibration of the sample, which is necessary for the accurate determination of electromechanical parameters.

In addition, according to the piezoelectricity standards (EN50324 [23] and IEEE Std. 176-1987 [24]), the parameters of the specimen or device under test (DUT) must be measured at a constant AC drive voltage or current. However, this criterion cannot be achieved with existing setups at high DC and AC fields due to the high output impedance. The problem can be illustrated even for simple AC small-signal constant voltage measurements, as depicted in Fig. 1. Here, a conventional piezoceramic sample is measured with a state-of-the-art impedance analyzer (Alpha-A, Novocontrol Technologies, Montabaur, Germany). It can be observed that the set voltage cannot be retained throughout the measurement and the drop at resonance frequency exceeds 80%. The sample is in series with the output impedance of the measurement system (here, the output impedance of the impedance analyzer is 50 Ω). The amplitude of the output signal is distributed proportionally to both impedances [25]. Since the impedance of the piezoceramic at resonance can be as low as a few ohms (orders of magnitude lower than the impedance of the measurement system), most of the amplitude of the output signal is lost through the output impedance of the standard measurement system, resulting in an AC signal amplitude of the sample that is lower than the set signal of the source. This issue is unfortunately often overlooked and can lead to significant errors in the determined material parameters even for small-signal measurements.

To overcome the above measurement problems and enable a detailed analysis of material parameters over a wide range of combined AC and DC signals, we have designed and implemented a new measurement system for piezoelectric resonance impedance spectroscopy under combined AC and high voltage (HV) DC loading. The proposed system is based on two separate output amplifier stages and incorporates an optimization algorithm to keep the AC conditions constant over the entire measurement frequency range, despite the highly nonlinear impedance curve. The functionality of the setup was verified using equivalent circuits and demonstrated by investigating the effect of large DC bias fields on the electromechanical properties of a ferroelectrically soft PZT. Small-signal electromechanical coefficients were determined by resonance impedance spectroscopy while a DC bias field of up to 5 kV/mm was applied along and opposite to the polarization direction between the poling field and the coercive field. In particular, the evolution of the resonance and antiresonance quality factors is followed and compared with the piezoelectric, mechanical, and dielectric coefficients ($d_{31}$, $k_{31}$, $s^{E}_{11}$, and $e^{T}_{33}$).

II. DESIGNED MEASUREMENT SYSTEM

A. Hardware

The designed measurement system enables measurements at constant AC voltage amplitude. The measurement capabilities and limitations are shown in Table I [26]. The main feature of the system (Fig. 2) is two separate output amplifier stages for the DC bias voltage and the AC signal, respectively. The DC voltage ($V_{\text{SDC}}$) is generated by the low-voltage DC source and amplified by the HV amplifier with a gain of 1000. To remove the high-frequency noise from the amplifier, the amplified DC voltage ($V_{\text{HV}}$) is filtered with the HV second-order passive low pass (LP) filter, before being fed to the DUT ($V_{\text{DUT(DC)}}$). An integrated HV DC voltmeter monitors the DC bias $V_{\text{DUT(DC)}}$.

The AC signal ($V_{\text{SAC}}$) is generated by the AC source. To increase the current capability, lower the output impedance, and protect the AC source in case of dielectric breakdown of the DUT, an additional analog buffer (high current broadband amplifier with a gain of 1) is used. An HV capacitor (DC blocking capacitor $C_1$) is used to protect the analog buffer from the HV DC bias voltage $V_{\text{DUT(DC)}}$. Capacitor $C_1$ acts as a high pass (HP) filter for the AC signal. Since the
The impedance of the DUT is calculated as the ratio of two phasors: measured AC signal $V_{DUT(AC)}$ and the current $I_{DUT(AC)}$ [28]. An HV HP filter removes the DC component of the voltage $V_{DUT(DC+AC)}$. The DUT AC current $I_{DUT(AC)}$ is measured with the current meter, which has four adjustable current ranges, i.e., three shunt resistors and a current probe. In this way, current measurement over several orders of magnitude is achieved. In addition, the laser vibrometer is used to measure the displacement of the DUT, which is caused by electromechanical coupling. The system is controlled by software (see Section II-B) that manages all components of the measurement system, i.e., data acquisition, current measurement, and AC and DC sources.

In case of dielectric breakdown of the DUT during DC application, the voltage of the DC blocking capacitor $V_{DUT(DC)}$ is applied to the output of the analog buffer, which is detrimental. Furthermore, if the breakdown occurs during the frequency sweep, high current surges flow into the current measurement component, which can destroy parts of the current measurement circuits. Therefore, both components (analog buffer and current measurement, see Fig. 2) are equipped with overvoltage protection elements (diodes, gas arrestors, capacitors, and ferrite beads) and sacrificial hardware component (simple, low-cost printed circuit board with operational amplifier and passive elements) that do not affect the behavior of the DUT during regular operation. Sacrificial hardware components are replaced in case of destruction.

### B. Control System and Measurement Procedure

The measurement automation program controls the components of the system. It contains an automated measurement procedure, data acquisition and processing, an optimization algorithm to keep the AC signal voltage $V_{DUT(AC)}$ amplitude constant, dielectric breakdown reduction techniques, and a self-calibration and self-test procedure.

The flowchart of the automatic impedance spectroscopy measurement procedure is shown in Fig. 3. It should be noted that the measurement procedure is invoked after the DUT DC bias voltage $V_{DUT(DC)}$ is automatically ramped up to the set value, typically at a rate of 1.2 kV/min. In the beginning, the output impedance of the voltage generating side is equal to the combined impedance of the analog buffer and capacitor $C_1$ and the output impedance must be as low as possible, a high capacitance capacitor (120 nF) is used [25]. The low output impedance (compared to the commercial impedance analyzers) should allow impedance spectroscopy measurement under constant DUT AC signal $V_{DUT(AC)}$ conditions over a wider impedance range of the DUT. However, for the case of extremely low DUT resonance impedance, a special amplitude optimization algorithm was implemented in the control software (see Section II-B) to keep $V_{DUT(AC)}$ constant over the specified measurement range.

To prevent dielectric breakdown in air, the measurement system includes a holder in combination with a gas chamber and a protective atmosphere (usually SF$_6$ insulating gas [27], others possible) that allows the DUT to resonate without rigid boundary conditions (rigid fixation) and enables measurements at low pressures or overpressures.

The impedance of the DUT is calculated as the ratio of two phasors: measured AC signal $V_{DUT(AC)}$ and the current $I_{DUT(AC)}$ [28]. An HV HP filter removes the DC component of the voltage $V_{DUT(DC+AC)}$. The DUT AC current $I_{DUT(AC)}$ is measured with the current meter, which has four adjustable current ranges, i.e., three shunt resistors and a current probe. In this way, current measurement over several orders of magnitude is achieved. In addition, the laser vibrometer is used to measure the displacement of the DUT, which is caused by electromechanical coupling. The system is controlled by software (see Section II-B) that manages all components of the measurement system, i.e., data acquisition, current measurement, and AC and DC sources.

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Fig. 3. Flowchart of the automated impedance spectroscopy measurement procedure.

frequency of the AC source \( f_{AC[n]} \) \((n = 1, 2, \ldots, n_{\text{MAX}})\) is set by block A. As mentioned earlier, the DUT parameters must be measured under constant AC signal conditions, depending on the user’s measurement mode setting. This is achieved by the optimization algorithm in block B. First, the actual DUT AC signal voltage \( V_{\text{DUT(AC)}} \) amplitude is measured. Then, the optimization algorithm calculates the required amplitude of the AC source signal \( V_{\text{SAC}} \) to achieve the desired DUT AC signal voltage \( V_{\text{DUT(AC)}} \) amplitude. The optimization steps are repeated until a sufficiently accurate \( V_{\text{DUT(AC)}} \) amplitude is achieved. In practice, an actual \( V_{\text{DUT(AC)}} \) amplitude within 1% of the desired amplitude is sufficient for highly accurate measurement of the DUT impedance. Note that due to the nonlinear behavior of the DUT at resonance, the Newton–Rapson method [29] is used as the optimization algorithm for calculating the \( V_{\text{SAC}} \) amplitude.

A suitable current measurement range is then selected in block C. After the desired measurement conditions are achieved, data acquisition of the DUT is performed by block D, i.e., the DUT AC signal amplitude \( V_{\text{DUT(AC)}} \) and current \( I_{\text{DUT(AC)}} \), as well as the displacement of the DUT over time. Condition E determines whether all specified frequency points have been measured. If the condition is met, the loop is terminated. Otherwise, blocks A–D are called again.

The measurement data from blocks F–G are processed, recorded, and stored in a parallel loop. The lock-in method [30] is used to calculate the impedance of the DUT and achieve a high signal-to-noise ratio (SNR).

The control program also includes a self-calibration and self-test procedure. For this purpose, resistors with a low tolerance are used as DUT. All measurement data are corrected according to the values determined during the calibration procedure.

III. MEASUREMENT RESULTS AND DISCUSSION

A. Verification of the Setup Accuracy

The suitability of the proposed measurement setup was first verified by performing a small-signal impedance scan at constant voltage on the same soft PZT sample, as reported in Fig. 1. The result shown in Fig. 4 confirms that the setup provides a constant voltage over the whole frequency range under investigation, despite the significant impedance change of the sample from 11 to 18 800 \( \Omega \).

To verify the accuracy of the system, the impedance spectrum of a reference equivalent circuit was recorded and compared with a measurement using commercially available impedance analyzers (Fig. 5). The reference equivalent circuit is shown in the inset of Fig. 5(b) and contained a dynamic LCR branch in parallel with a static capacitance (tolerance ±5% for capacitance and ±10% for others), resembling a piezoelectric resonator [31].

The measurement results are in excellent agreement, confirming the measurement accuracy of the designed measurement system. Note that for this comparative study, equivalent circuits were chosen instead of piezoelectric materials to ensure reproducibility regardless of measurement history, aging time, or drive voltage. Furthermore, the superposition of the (moderate) DC voltage had no effect on the impedance characteristics of the equivalent circuit; thus, the measurement accuracy was confirmed even with combined AC and DC excitation. In addition, comparative measurements have been carried out using a soft piezoceramic PZT (PIC151). The calculated material parameters revealed an excellent agreement between setups (Supplementary Table SI).

B. Measurement of Soft PZT

Fig. 6 shows the resonance impedance spectra of a soft PZT (PIC151, PI Ceramic GmbH, Lederhose, Germany) in
the transverse length vibration mode [23] at three exemplary DC bias fields. The electromechanical small field coefficients calculated according to [23] are presented in Fig. 7, as a function of the applied DC bias field. Note that $Q_{31}^{R}$ and $Q_{31}^{AR}$ were determined using the 3 dB method. Positive and negative field values denote the field applied along and opposite to the polarization direction, respectively. The descending branch of the DC bias field is shown. The mechanical quality factors in resonance ($Q_{31}^{R}$) and antiresonance ($Q_{31}^{AR}$) increase continuously and almost linearly with the increase of DC bias voltage. Dielectric breakdown at 5.5 kV/mm occurs before the onset of $Q_{31}^{AR}$ saturation. For negative DC bias fields, the $Q_{31}^{R/AR}$ values decrease and drop to zero near the coercive field (1.1 kV/mm, polarization and strain hysteresis loops are given in Supplemental Fig. S1 for comparison), which is characterized by the disappearance of the resonance/antiresonance peaks. The piezoelectric coefficient ($d_{31}$), electromechanical coupling coefficient ($k_{31}$), elastic compliance ($s_{11}^{E}$), and dielectric permittivity ($\varepsilon_{31}^{E}$) exhibit the opposite DC bias field dependence [Fig. 7(b)–(e)]. These coefficients decrease continuously with increasing DC bias field along the polarization direction, which is consistent with previous reports [18], [19], [21]. The slopes decrease at high fields, indicating the onset of saturation. At low fields, the coefficients increase but fall rapidly toward zero near the coercive field.

The decrease in piezoelectric coefficients and the increase of $Q_{31}^{R/AR}$ at positive DC bias field indicate ferroelectric hardening; vice versa, the increase of the coefficients and the decrease of $Q_{31}^{AR}$ at negative DC bias field indicate ferroelectric softening [32]. The coefficients vanish at the coercive field due to depolarization caused by an electric field. It should be noted that the calculation of the coefficients takes into account the geometry of the sample and the static strain induced by the DC bias field must be considered. For example, to take into account the change in the sample length induced by the DC bias field ($E_{bias}$), the calculation of elastic compliance [23] is modified as follows:

$$s_{11}^{E} = \left(4\rho f_{R}^{2}/l_{0}\right)^{-1} = \left(4\rho f_{R}^{2}(l_{0} + l_{0}d_{31}E_{DC})^{2}\right)^{-1} \quad (1)$$

where $l_{0}$ denotes the initial sample length and $l$ denotes the length at DC bias. $\rho$ and $f_{R}$ are the resonator density and the determined resonance frequency, respectively. However, the influence of static strain on the coefficients is in the range of 1% and can usually be neglected.

C. Origin of the Quality Factor Change With DC Bias Field

Resonance impedance spectroscopy at DC bias field can be rationalized using the following considerations. The applied DC bias field can be considered as a point at the polarization and strain hysteresis loops of the sample in the polarized and aged state, at which a small AC electric field and a resulting dynamic mechanical stress are superimposed. The reduction of the Gibbs free energy for polarization directions along the electric field (Landau–Ginsburg–Devonshire theory [33], [34]) forces an elimination of all domains with unfavorable polarization vectors, leading to a growth of the domain size and reduction of domain wall density. Moreover, the DC bias field will reduce domain wall mobility by stabilizing the newly formed domain configuration and steepening the energy landscape [2]. In a simplified representation, this can be described as an electric field-induced transition from a Rayleigh-like to a V-potential energy landscape [12], [35], [36].

When an additional dynamic perturbation is applied, i.e., a small AC electric field or the resulting mechanical stress, the coefficients $e = \partial P/\partial E$, $s = \partial S/\partial T$, ...
of irreversible domain wall motion. An alternative explanation arises from the facts that the applied DC bias fields are well above the composition’s coercive and poling fields (2 kV/mm) and that the $d_{31}$, $k_{31}$, $s_{11}^{T}$, and $\varepsilon_{33}^{T}$ values indicate a beginning of saturation. Both suggest that the compositional electromechanical coefficients and quality factors in the region of the maximum positive DC bias field might be dominated by the intrinsic contributions. The origin of the intrinsic loss is likely the susceptibility of the composition to lattice polarization rotation, as has been demonstrated in single domain single crystals [4]. A small ferroelectric crystal lattice distortion is correlated with a shallow Gibbs free energy distribution according to the Landau–Ginsburg–Devonshire theory [33], [34]. The flat energy distribution facilitates the extension of the electric dipole in the lattice and promotes the rotation of the lattice polarization vector upon external perturbation [37], which remains to be confirmed by further investigations.

At negative DC bias fields, two processes are coupled. At low field values, the reverse DC bias field effect occurs, i.e., the field opposite to the polarization direction destabilizes the domain configuration, facilitating (irreversible) domain wall motion and leading to enhanced $d_{31}$, $k_{31}$, $s_{11}^{T}$, and $\varepsilon_{33}^{T}$ and reduced $Q_{31}^{R/AR}$ values. Upon reaching the coercive field, the sample depolarizes rapidly.

**D. Impact of Experimental Conditions on the DC Bias Dependence of the Electromechanical Coefficients**

Most PZT-based compositions are prone to a severe decrease of $Q_{31}^{R/AR}$ upon increasing vibration velocity [5], [38], [39], which is also a potential error source for the small-field measurements under DC bias. Resonance impedance spectroscopy is usually conducted under constant driving AC electric field amplitudes. The maximum vibration velocity occurs at resonance and is determined by the applied field amplitude and the electromechanical coefficients [40]

\[
\nu_i = \frac{4}{\pi} \sqrt{\frac{\varepsilon_{ij}^{T}}{\rho}} \cdot k_{ij} \cdot Q_{ij} \cdot E_j = \frac{4}{\pi} \sqrt{\frac{d_{ij}}{\varepsilon_{ij}^{T}} \cdot Q_{ij} \cdot E_j}. \quad (2)
\]

Suppose that the electromechanical coefficients undergo a change due to an external perturbation, e.g., a DC bias field. Then, the driving field amplitude must be adjusted to maintain a constant (resonance) vibration velocity between measurements. Therefore, the application of the constant field measurement conditions induces different vibration states, which can have an enormous impact on the measured $Q_{31}^{R}$ values, due to the substantial change of $Q_{31}^{R}$ as a function of vibration velocity (especially in the range of low vibration velocities).

Fig. 8 schematically displays the situation of a strong $Q_{31}^{R}$ decrease as found in PZT compositions [41]. An external parameter (DC bias field) will predominantly change the $Q_{31}^{R}$ values but not the stability of the vibration velocity [13]. Therefore, the measurement conditions with constant field amplitude or constant vibration velocity lead to different $Q_{31}^{R}$ values for different external parameters. To overcome this problem, we propose to perform the measurements with a constant resonance vibration velocity (as was done in Figs. 6 and 7).

and $d = \partial S / \partial E = \partial P / \partial T$ are determined by the slope of the polarization/strain hysteresis loop at the point employed by the DC bias field (neglecting the frequency difference between large- and small-signal loops). The slope and hence the coefficients decrease with increasing DC bias field as a consequence of the immobilization of the domain walls. Moreover, the hysteretic character of the dynamic response decreases for large DC bias fields, implying reduced irreversible domain wall motion [20]. Since the irreversible motion of the domain walls is considered to be the dominant source of loss under resonance vibration, the decrease at large DC bias fields is associated with an increase in the mechanical quality factors $Q_{31}^{R/AR}$. However, the $Q_{31}^{R/AR}$ values remain comparatively small compared to ferroelectrically hard PZT compositions [5]. The small values are related to the flat potential and the absence of acceptor-based domain wall pinning, which still allows for a substantial residual amount

Fig. 7. Electromechanical coefficients (a) $Q_{31}^{R}$ (solid symbols) and $Q_{31}^{AR}$ (open symbols), (b) $d_{31}$, (c) $k_{31}$, (d) $s_{11}^{T}$, and (e) $\varepsilon_{33}^{T}$ in the transverse (31) vibration mode as a function of DC bias field ($E_{bias}$). The measurements were conducted at a vibration velocity of 0.01 m/s (0.14–0.17 V/mm AC excitation voltage) and 20 °C. Positive and negative field values denote the field applied along and opposite to the polarization direction, respectively.

\[ v_i = \frac{4}{\pi} \sqrt{\frac{\varepsilon_{ij}^{T}}{\rho}} \cdot k_{ij} \cdot Q_{ij} \cdot E_j = \frac{4}{\pi} \sqrt{\frac{d_{ij}}{\varepsilon_{ij}^{T}} \cdot Q_{ij} \cdot E_j}. \quad (2) \]
The effect concerns mainly the setting the DC bias field and ranged from 0.14 to 0.17 V/mm. The applied DC bias influences the vibrations to external perturbations. The difference between conditions in the design and construction of the measurement setup and design enables to carry out piezoelectric resonance impedance spectroscopy at high DC bias fields, complying with established standards, ensuring appropriate protection of the device, DUT, and the user. Functionality of the proposed setup was benchmarked against commercial impedance analyzers in the small-signal range and reference equivalent circuits. Testing was carried out by determining the electromechanical parameters of a soft PZT composition. Results revealed a more substantial decrease of piezoelectric coefficients upon applying constant AC voltage drive stability in (Na$_{1-x}$Bi$_x$)$_2$TiO$_3$ and BaTiO$_3$ based piezoceramics, Acta Mater., vol. 227, Apr. 2022, Art. no. 117703.

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