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The effect of electric field-induced phase transitions on the blocking force in lead-free ferroelectrics

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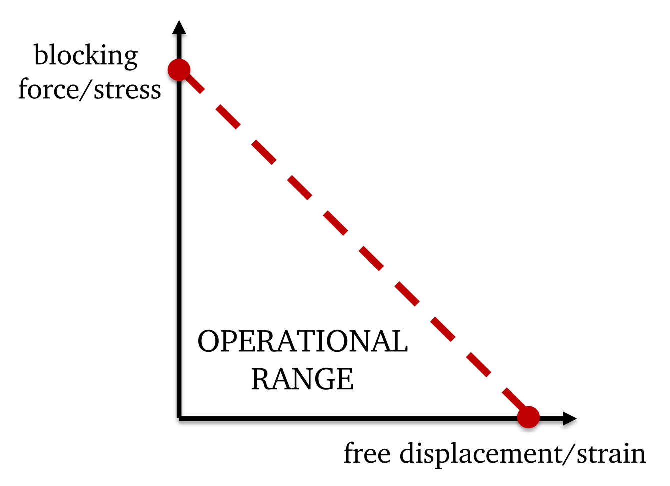
Project D6

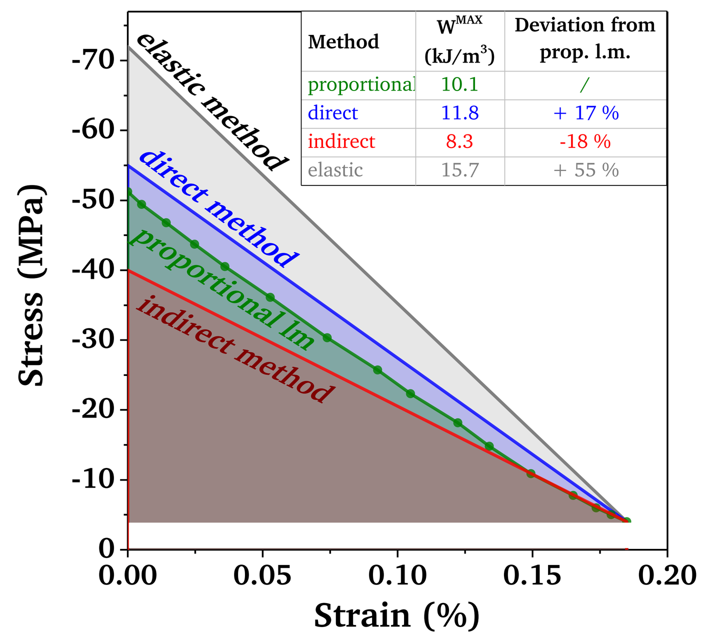
Motivation

1) Methodology and equipment development

- Review the currently used methodology and identify problems.
- Develop new methodology and required measurement equipment.
- Test new methodology on lead-based piezoelectrics and compare to other methods.



Operational range of piezoelectric actuators is typically determined by linear interpolation between free strain (max. strain in mechanically unloaded conditions) and blocking stress (max. stress in fully clamped state). The latter can be either measured (direct and indirect method) or extrapolated (elastic method). [1]



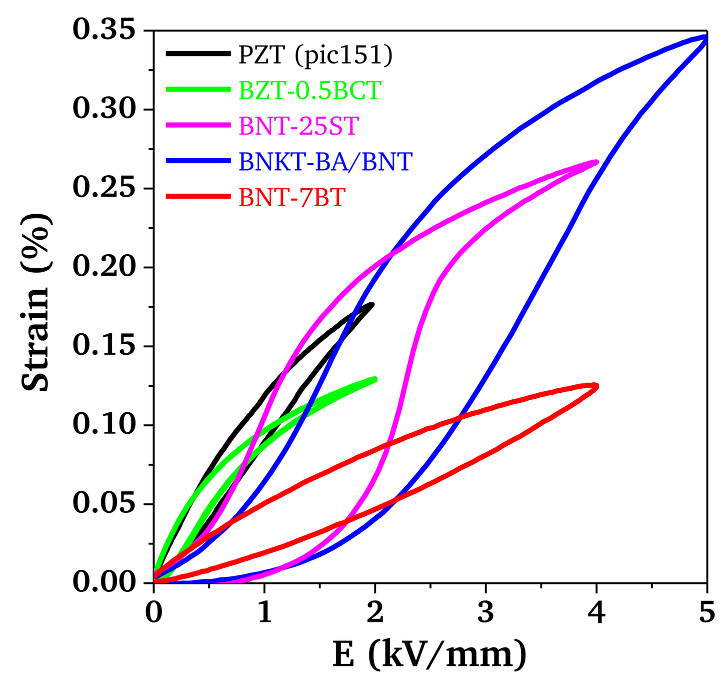
Method	W^{MAX} (kJ/m^3)	Deviation from prop. l.m.
proportional	10.1	/
direct	11.8	+ 17 %
indirect	8.3	-18 %
elastic	15.7	+ 55 %

Operational range of soft PZT ceramics at E_{max}= 2 kV/mm, as determined by different methods. The large differences originate from the inherent hysteretic electromechanical behavior of ferroelectrics and the different order of application of the electrical and mechanical loads by each method. [4] The inset table compares the calculated maximum work outputs.

2) Operational range of lead-free piezoelectrics

Many lead-free piezoelectrics display exceptional large electromechanical properties that are in some cases larger than the current state-of-the-art lead-containing materials. The goals of the second part were therefore to:

- Investigate the operational range of lead-free piezoelectrics and compare it to the established lead-based piezomaterials.
- Investigate the influence of the electric-field-induced transitions on the operational range and compare it to other force developing mechanisms in ferroelectrics.



Unipolar strain of some promising lead-free compositions:

- *Ferroelectric Ba($Zr_{0.2}Ti_{0.8}$)O_3-($Ba_{0.7}Ca_{0.3}$)TiO_3 (BZT-BCT)*
- *Relaxor ferroelectric (ergodic) 0.75($Bi_{1/2}Na_{1/2}$)TiO_3-0.25$SrTiO_3$ (BNT-25ST)*
- *Relaxor ferroelectric (non-ergodic) 0.93($Bi_{1/2}Na_{1/2}$)TiO_3-0.07$BaTiO_3$ (BNT-7BT)*
- *Relaxor/Ferroelectric composites $Bi_{1/2}$($Na_{0.75}K_{0.25}$)$_{1/2}TiO_3$-$BiAlO_3$/($Bi_{1/2}Na_{1/2}$)TiO_3 (BNKT-BA/BNT)*

Soft PZT (pic151, PI Ceramics) is added for comparison.

3) Industrial multilayer actuators (work in progress)

- Use the knowledge obtained from 1) and 2) to investigate the operational range of industrial multilayer actuators and determine the influence of the multilayer structure on the mechanical and electrical constitutive behavior.

Key Publications (2013-2014)

1) WEBBER K., FRANZBACH D. J., and KORUZA J. Determination of the true operational range of a piezoelectric actuator. *Journal of the American Ceramic Society*, doi: 10.1111/jace.13024 (in press), 2014.

2) BRANDT D. R. J., ACOSTA M., KORUZA J., and WEBBER K. Mechanical constitutive behavior and exceptional blocking force of lead-free BZT-xBCT piezoceramics. *Journal of Applied Physics* 115(20): 204107, 2014.

3) DANIEL L., HALL D. A., WEBBER K. G., KING A., and WITHERS P. J., Identification of crystalline elastic anisotropy in PZT ceramics from in-situ blocking stress measurements. *Journal of Applied Physics* 115: 174102, 2014.

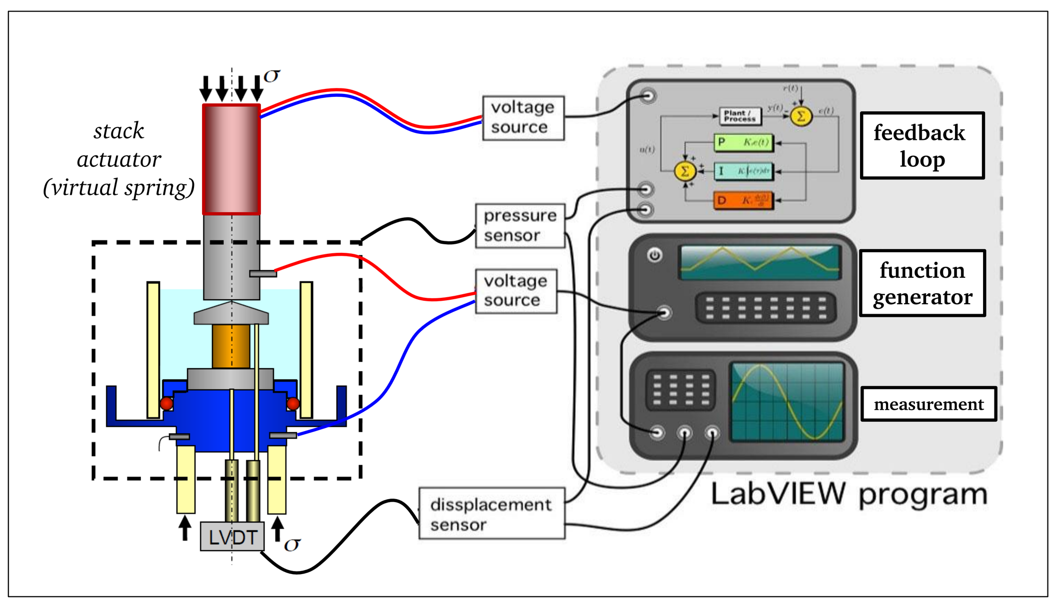
4) KORUZA J., FRANZBACH D., SCHADER F., ROJAS V., and WEBBER, K. Enhancing the Operational Range of Piezoactuators by Uniaxial Compressive Preloading. *Journal of Applied Physics* (submitted), 2014.

5) DANIEL L., HALL D. A., KORUZA J., WEBBER K. G., KING A., and WITHERS P. J. Revisiting the blocking force experiment on ferroelectric ceramics using high energy x-ray diffraction. *Journal of the European Ceramic Society* (submitted), 2014.

Note: The Project D6 started in January 2013.

Results

1) Methodology and equipment development

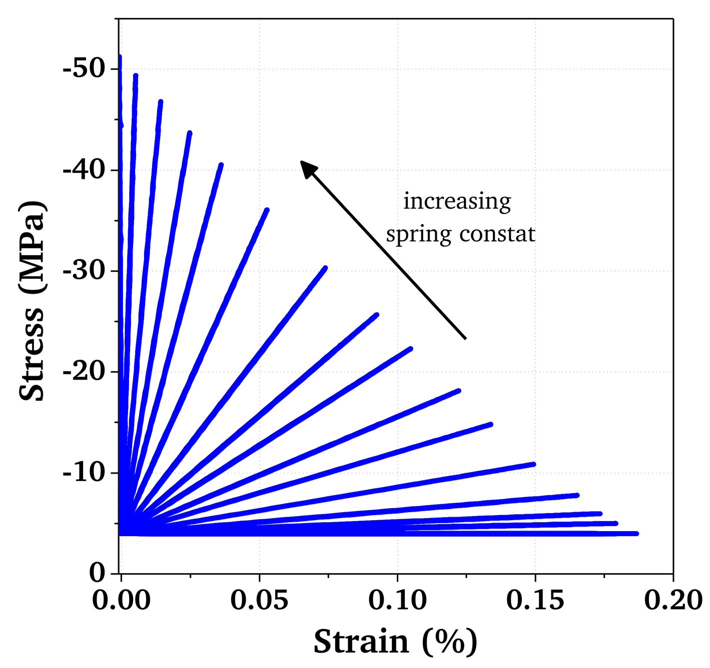


The newly developed setup for the determination of the operational range by proportional loading, which enables the simulation of real application conditions.

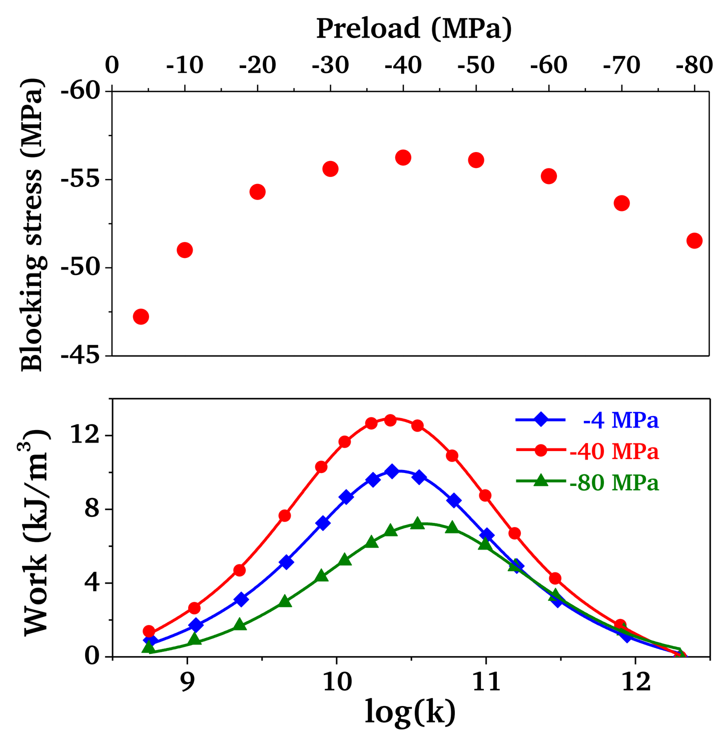
Measurement capabilities:

- *Compressive preloads (1 – 400 MPa)*
- *Temperatures (20 – 200 °C)*
- *Frequencies up to 50 mHz*
- *Simulation of external loads with different linear or non-linear stiffnesses*

Unipolar electric field is applied to the sample while the external load with a predefined spring constant is simulated by a large stack actuator (virtual spring), controlled by a PID feedback loop. The force and displacement are detected by a force sensor and an LVDT, respectively.

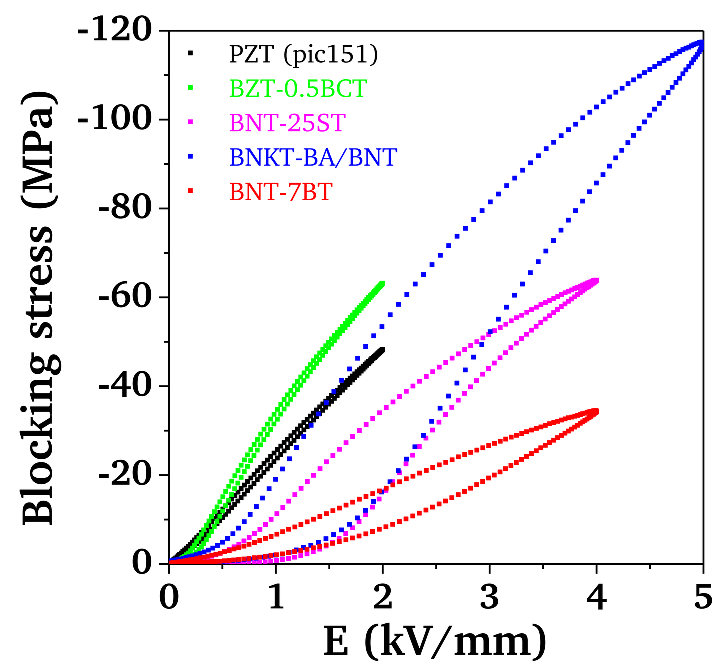


The full stress-strain operational range of soft PZT, as measured by the proportional loading method at a compressive preload of -4 MPa and an E_{max} of 2 kV/mm. [1]

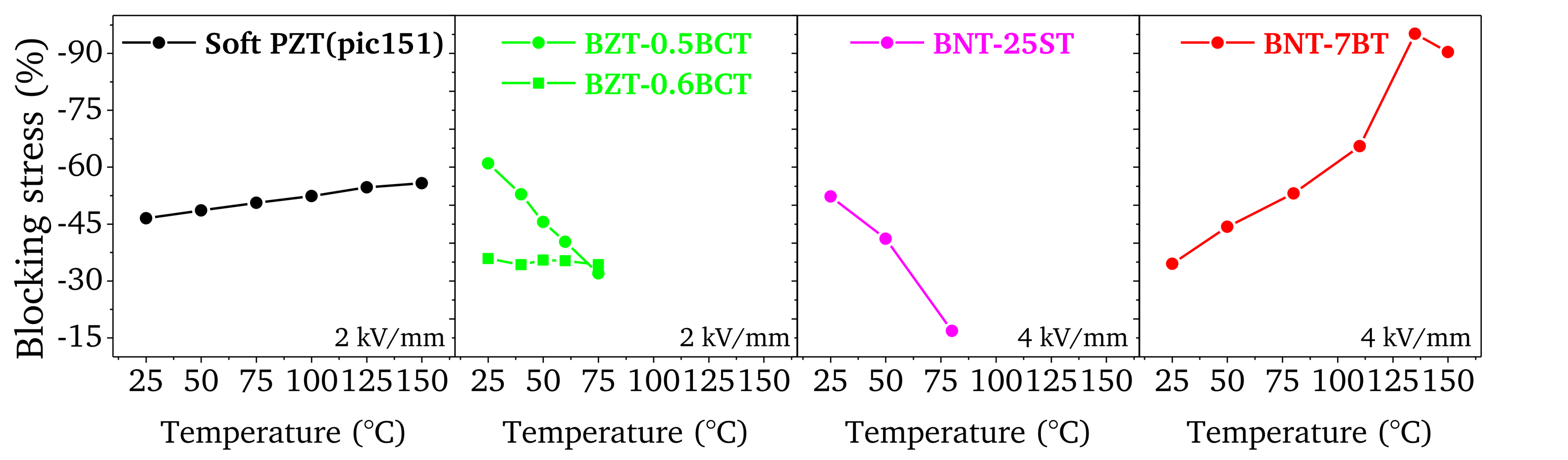


Dependence of the blocking stress on the uniaxial preload (above) and work as a function of the applied spring constant for selected values of the preload (below) for PZT piezoceramics (2 kV/mm). [4]

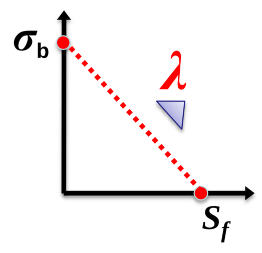
2) Operational range of lead-free piezoelectrics

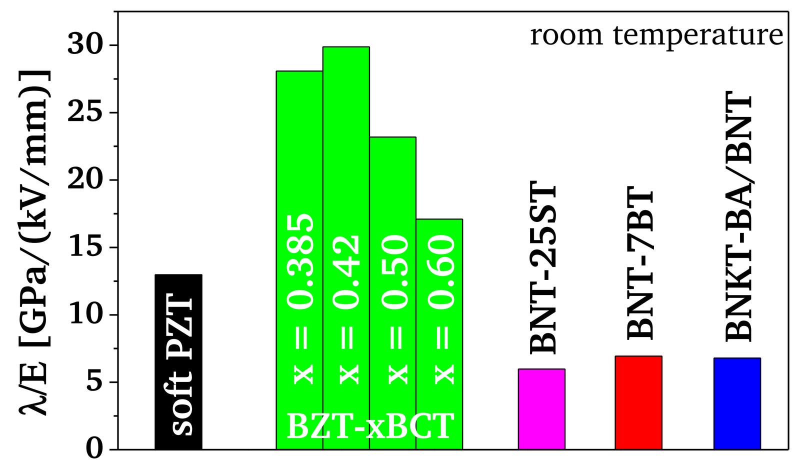


Electric field dependent blocking stress of various lead-free piezoelectrics at room temperature, compared to soft PZT. Large forces, developed by BNT-25ST and BNKT-BA/BNT can be related to the reversible electric-field-induced phase transition and the composite effect, respectively. On the other hand, BZT-0.5BCT exhibits high blocking force values and linear behavior at lower electric fields. [3]



Temperature dependence of the blocking force of various lead-free materials in comparison with soft PZT.





The electromechanical conversion efficiency λ is defined as the amount of the free displacement that can be converted into force in the fully clamped state, and can be calculated as the secant modulus connecting the free strain and the blocking stress (schematics left). For comparison between different materials the values were normalized by the applied electric field.