Journal of Electroceramics, 15, 57–64, 2005

Poling Conditions of Pre-Stressed Piezoelectric Actuators and Their Displacement

J. JUUTI,* H. JANTUNEN, V.-P. MOILANEN & S. LEPPÄVUORI

Microelectronics and Materials Physics Laboratories, Department of Electrical and Information Engineering, EMPART Research Group of Infotech Oulu, University of Oulu, Finland, P.O. Box 4500, FIN-90570 Oulu

Submitted August 10, 2004; Revised January 15, 2005; Accepted March 22, 2005

Abstract. The poling procedure has always been the key issue in producing piezoelectric actuators with optimised performance. This is also true with the relatively new category of pre-stressed bender actuators, where mechanical bias achieved with a passive layer is introduced in the actuators during manufacturing. Due to these factors, the behaviour of the actuator under poling is different compared to its bulk counterparts. In this paper, two different thicknesses of commercial PZT 5A and PZT 5H materials were used in bulk actuators and pre-stressed benders realised by new method. Pre-stress was introduced by using a post-fired biasing layer utilising sintering shrinkage and difference in thermal expansion. The hysteresis loop of the actuators was measured under 0.5–7.0 MV/m electric fields at 25–125°C temperatures, providing information about their remnant polarisation and coercive field before poling. The results showed that high electric field and 25°C temperatures in poling provided higher remnant polarisation and coercive electric field than using 125°C temperature at poling. Difference was especially significant in coercive electric field values where up to 114.8% difference was obtained for PZT 5H bulk actuator and 65.9% for pre-stressed actuators. Higher coercive fields can be utilized as increased operating voltage range of piezoelectric devices. The differences in results obtained here and by others can be explained by the different pre-stress level, stronger clamping of the thicker passive layers of the RAINBOW and THUNDER actuators and passive ring area introducing high tensile stresses. The same conditions were used to pole the actuators, after which the displacement and dielectric constant of the actuators were measured. The displacement measurements showed that remnant polarisation has good correlation with displacement. This fact can be used in estimating pre-stressed actuator performance before actual poling. The dielectric constant measurements with a small signal after poling gave even better correlation than the remnant polarisation.

Keywords: piezoelectric ceramic, pre-stress, actuator, poling

1. Introduction

To obtain higher mechanical output power and efficiency, piezoelectric actuators are currently operated under increased stresses/electric fields. Therefore, it has become increasingly important to control the whole preparation procedure of the actuators and to characterise the dielectric properties of the piezoelectric material under actual conditions. Although the compositions and process parameters used, as well as final ageing, are important, optimisation of the poling process is one of the key issues in producing actuators with maximum performance [1, 2]. In the poling process, the actuator acquires its actual piezoelectric properties; the shape of the hysteresis loop, permittivity, dielectric loss and displacement properties at high electric field driving. These properties are weakened afterwards only to some extent by ageing [3–5]. It is therefore natural that the poling conditions of planar actuators have been widely studied and, e.g., Wang et al. have shown that optimised poling conditions of bulk actuators can be developed through predetermination of saturation polarisation and the coercive field [6].

*To whom all correspondence should be addressed. E-mail: jajuu@ee.oulu.fi

Pre-stressed actuators are bending actuators having internal stress introduced in their manufacturing process. The common way to prepare them is to use two different layers with different thermal expansions and/or sintering shrinkages [2, 7–9]. Thus, the achieved unimorph structure bends, and the internal pre-stress acts as a spring. The produced components can exhibit large displacements due their higher coercive electric field and improved piezoelectric coefficients affected by their internal stress [3, 10–12]. It is therefore natural that the properties and methods of producing pre-stressed actuators are extensively investigated [13–16]. However, only little has been reported about the optimisation of the poling conditions.

The main purpose of this paper is to study the optimisation of the poling conditions of pre-stressed piezoelectric actuators of two different thickness and two different materials. The results are compared to properties of the bulk actuators with same geometry. Properties of the actuators achieved here are also compared to results obtained by others with other pre-stressing methods (RAINBOW and THUNDER). Optimisation is done firstly by measuring their remnant polarisation and coercive field before poling and secondly, by measuring their displacement and dielectric constant after different poling conditions. Remnant polarisation gives the polarisation level achieved by poling due to domain orientation. The coercive electric field of the hysteresis loop, i.e., the electric field that needs to be applied to the components in order to get zero remaining charge, is used because it specifies the usable electric fields for component excitation. The method is basically the same as the one introduced by Wang et al. [17] for non-pre-stressed actuators. The displacement and dielectric constant are used to verify the final properties.

2. Experiments

Commercial piezoelectric PZT 5A and PZT 5H (Morgan Electro Ceramics) bulk materials were selected for the pre-stressed actuators, as was also done in many other studies of related structures manufactured by different methods [9, 12]. Pre-stressing was introduced as a mechanical stress created by a post-fired metal layer during firing process, method is presented more detailed elsewhere [18]. The piezoceramic samples were 25 mm in diameter and 0.375 mm or 0.25 mm thick without electrodes. Commercial silver paste was used for the electrodes. A different AgPd paste was used for pre-stressing. The emulsion thickness of the screen-printing mask was 20 μm for the electrodes and 60 μm for the electrode/pre-stressing layer. The mask was circular in shape with a 24.5 mm diameter to prevent paste from going over the edges of the sample. This procedure also produces a circular insulation area, enabling a higher electric field without sparking on the edges. Bulk and pre-stressed samples were made the same way, but in the case of the pre-stressed actuators, the bottom electrode was a pre-stressing layer. After screen-printing and drying the electrodes, the discs were fired at 850°C in a belt furnace. The fired samples with a thickness of 0.25 mm were saddle or mixed-shaped, depending on the material, and the samples with a thickness of 0.375 mm were dome-shaped.

After a 24 h period had elapsed from the firing, the hysteresis loop was determined using temperatures in a range of 25–125°C and an electric field of 0.5–7.0 MV/m, depending on the component thickness and material. The samples were poled with a 30 min poling time using the same temperatures and electric fields as in the hysteresis loop measurements, and the dielectric constant and displacement characteristics were measured 24 h after poling. Furthermore, the actuators were aged for 10^5 cycles and the dielectric and displacement properties were measured again.

The hysteresis loop, remnant polarisation, $\pm P_r$, and coercive field, $\pm E_c$, measurements were carried out with a Radiant RTV6000HVS system (Radiant technologies, USA) and an HP 4284A Precision LCR meter was used with 1 V_{rms} and 1 kHz for the dielectric constant measurements. Displacement was measured with a system based on a Michelson interferometer [19]. An electric field of ±0.15 MV/m, 10 Hz and 0.3 N point load was used in the measurements.

3. Results and Discussion

The representative remnant polarisations and coercive fields as a function of temperature and electric field for different materials and thickness combinations as well as for pre-stressed and non-pre-stressed (bulk) structures are shown in Figs. 1–5. An important general result is that, with only one exception (250 μm thick pre-stressed PZT 5A actuator, Fig. 1), the $\pm P_r$ and $\pm E_c$ values of all the samples were highest at a 25°C temperature (Table 1). Thus, for the studied structures, thickness and materials, it is more advantageous to use

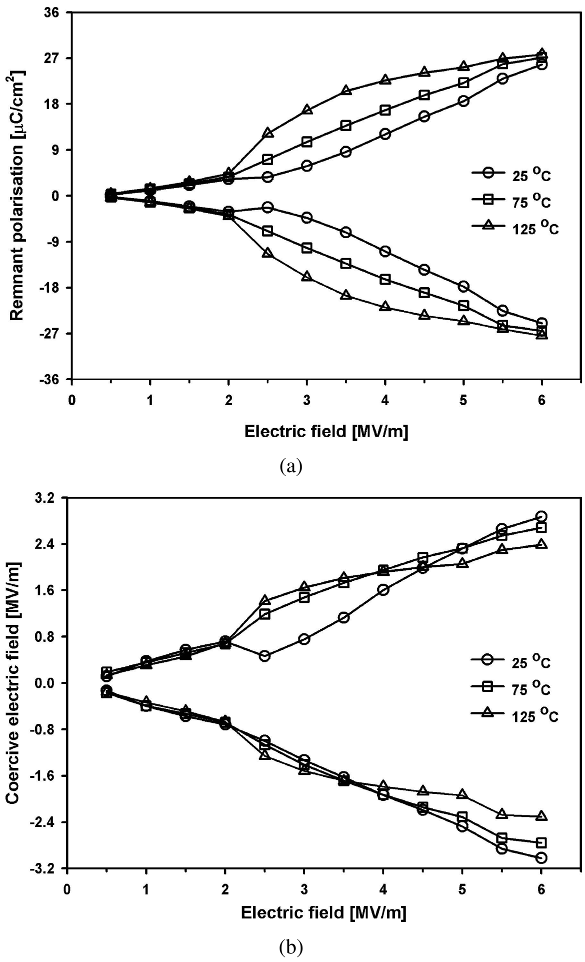


Fig. 1. (a) Remnant polarisation. (b) Coercive electric field of the pre-stressed PZT 5A actuator (thickness 250 μm) as a function of electric field and temperature.

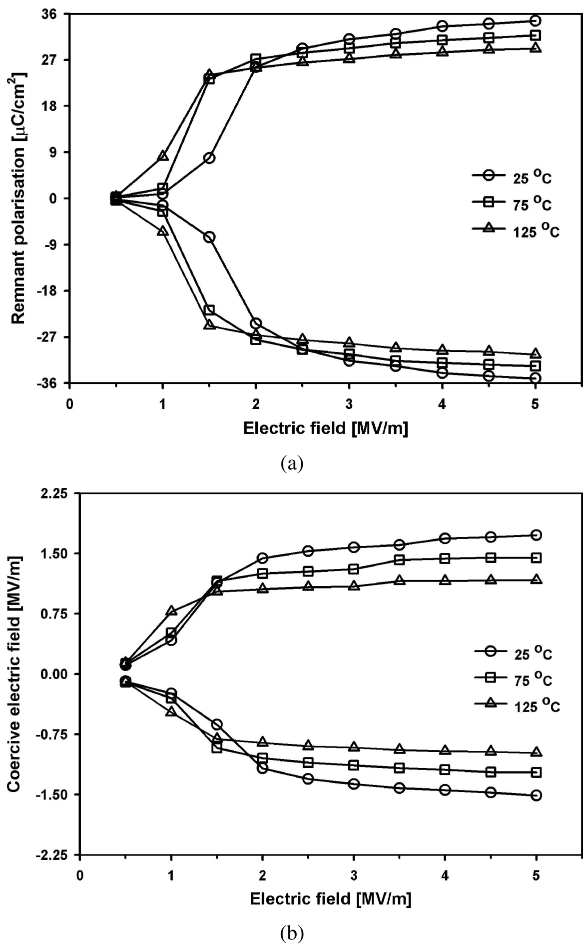


Fig. 2. (a) Remnant polarisation. (b) Coercive electric field of the PZT 5A bulk actuator (thickness 375 μm) as a function of electric field and temperature.

a high electric field than high temperature in poling, which agrees well with the conclusion reported earlier for RAINBOW (Reduced And INternally Biased Oxide Wafer) and THUNDER (THin layer composite UNimorph ferroelectric DrivER and sensor) actuators constructed from the same materials [3, 11].

Figures 2 and 3 shows typical result that poling of a pre-stressed actuator must be optimised more carefully than that of bulk counterparts, because the remnant polarisation and coercive field values of the pre-stressed actuator depend more gradually on the poling field than in the case of the bulk counterparts, which achieve their saturated values quickly with increased electric field. This is due the reason that bulk actuators are more free to shrink/expand to near their maximum level during poling since constraining layer is not existing. This is also true though active material is constrained by the ring of passive material (insulation area) generating large tensile and shear stresses. Stresses in the bulk actuator were modelled with ATILA software using material parameters of the manufacturer (Table 2).

In order to verify hindering effect of the ring in poling, passive ring area was coated with conductive silver paint making it active. The hysteresis measurements were repeated using the same parameters as earlier. Due the modification remnant polarization was typically slightly increased and coercive electric field was significantly increased (Fig. 4). Sparking on the edges prevented usage of the highest electric fields in the measurements and break some of the samples.

Table 1. Hysteresis measurement results under the highest electric fields.

Sample	Thickness (μm)	$+P_r$ (μC/cm^2) (25°C/125°C)	$-P_r$ (μC/cm^2) (25°C/125°C)	$+E_c$ (MV/m) (25°C/125°C)	$-E_c$ (MV/m) (25°C/125°C)
PZT 5A bulk	250	34.86/29.60	−35.61/−30.05	1.84/1.19	−1.67/−1.08
PZT 5A pre-stressed	250	25.77/27.72	−25.02/−27.42	2.87/2.38	−3.02/−2.31
PZT 5A bulk	375	34.63/29.22	−35.08/−30.42	1.73/1.17	−1.51/−0.98
PZT 5A pre-stressed	375	33.50/29.75	−32.90/−29.60	1.99/1.43	−2.23/−1.44
PZT 5H bulk	250	27.87/19.46	−29.07/−20.66	1.11/0.57	−1.05/−0.55
PZT 5H pre-stressed	250	21.33/20.21	−22.69/−20.36	2.04/1.23	−1.93/−1.17
PZT 5H bulk	375	29.30/21.18	−29.30/−21.33	1.16/0.54	−1.15/−0.52
PZT 5H pre−stressed	375	23.21/19.61	−22.16/−18.56	1.60/1.08	−1.77/−1.08

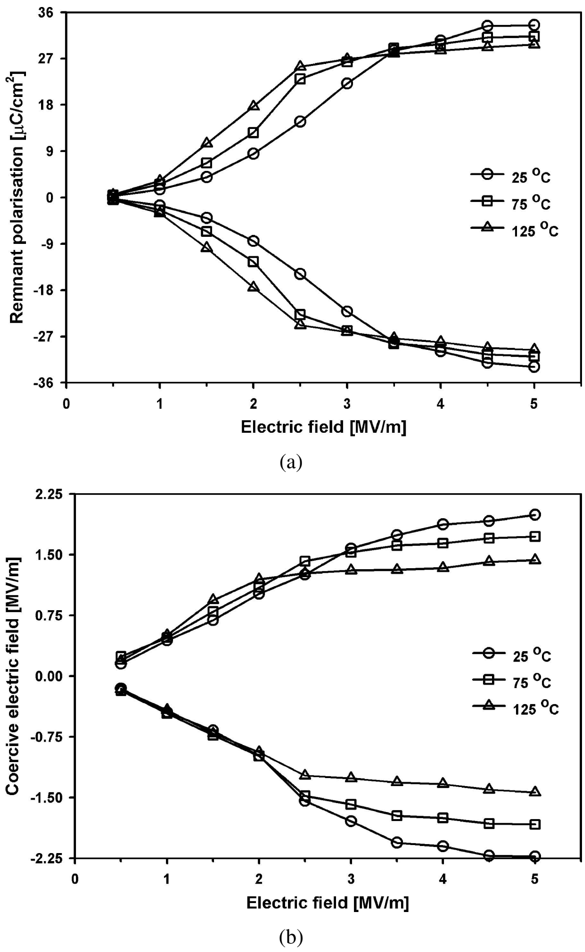


Fig. 3. (a) Remnant polarisation (b) Coercive electric field of the pre-stressed PZT 5A actuator (thickness 375 μm) as a function of electric field and temperature.

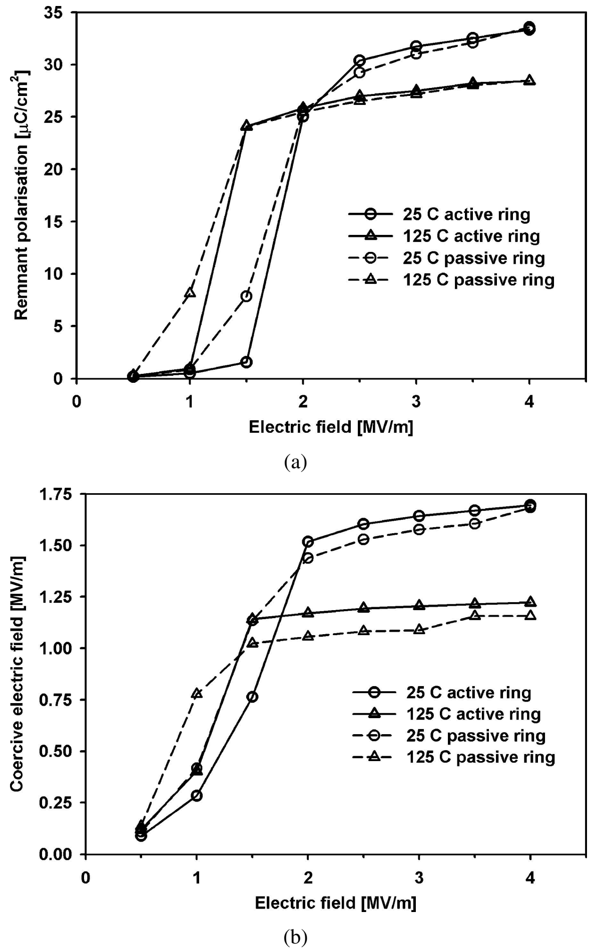
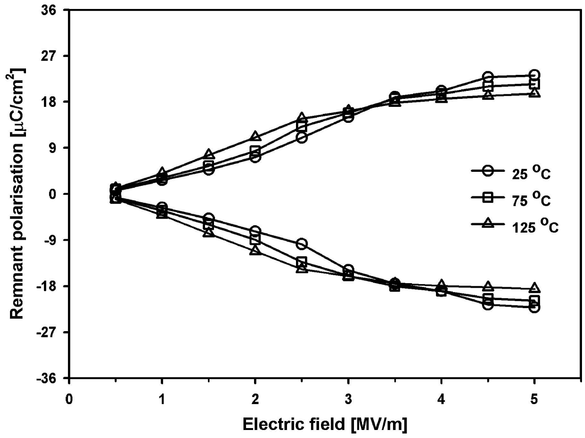
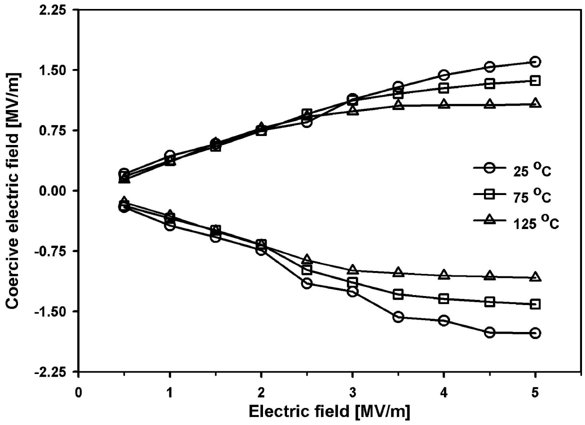


Fig. 4. (a) Remnant polarisation. (b) Coercive electric field of the bulk PZT 5A actuator (thickness 375 μm) as a function of electric field and temperature with passive and active ring.



(a)



(b)

Fig. 5. (a) Remnant polarisation. (b) Coercive electric field of the pre-stressed PZT 5H actuator (thickness 375 μm) as a function of electric field and temperature.

Same behaviour but lower stresses are expected to occur with pre-stressed actuators, due smaller in-plane displacement of the benders. To avoid decreased polarisation, actuators should be manufactured with full electrodes, yet effect of the stresses on displacements are not known.

In the general category of the results goes also the result that the PZT 5H material had 9–37% lower remnant polarisation and 20–54% lower coercive electric field values than the PZT 5A material (Table 1, Figs. 3 and 5). This result is well in line with the results reported earlier where 31–33% difference in remnant polarisation and 45–50% difference in coercive electric field was measured by Dausch [3]. Although, mentioned characteristics of PZT 5H are lower it has higher d_{31} piezoelectric

Table 2. Stresses of the bulk actuators modelled with ATILA software under the highest electric fields.

Sample	Thickness (μm)	Max stress due in-plane displacement (MPa)	Max stress due out-of-plane displacement (MPa)
PZT 5A, bulk	250	41	41
PZT 5A, bulk	375	30	42
PZT 5H, bulk	250	74	68
PZT 5H, bulk	375	50	64

coefficients (PZT 5H: $d_{31} = -274$ pm/V and PZT 5A: $d_{31} = -171$ pm/V, manufacture's information), which is generally preferable for large-displacement bending actuators.

In more detail, the pre-stressed actuators showed a significantly higher coercive electric field than their bulk counterparts, but decreased remnant polarisation (Table 1). The highest remnant polarisation for the bulk and pre-stressed actuators were almost the same (33.5 μC/cm^2 for the pre-stressed 375 μm thick PZT 5A actuators and 34.6 μC/cm^2 for the bulk counterpart in Figs. 2 and 4). Dausch reports opposite behaviour of remnant polarisation between RAINBOW and bulk actuators, 18.0 μC/cm^2 and 14.1 μC/cm^2 for PZT 5H actuators and 26.1 μC/cm^2 and 21.0 μC/cm^2 for PZT 5A, respectively [3]. Ounaies et al. reports nearly equal remnant polarisation for PZT 5A bulk and THUNDER actuators ($+P_{r\text{THUNDER}}$ = 34–35 μC/cm^2, $-P_{r\text{THUNDER}}$ = -32.5 to -34, $\pm P_{r\text{bulk}} = \pm 34$ μC/cm^2) [11]. Explanation can be higher pre-stress of RAINBOW actuators increasing stress induced domain reorientation [9, 20].

The highest coercive field was achieved with pre-stressed 250 μm thick PZT 5A actuators, 2.87 MV/m and -3.02 MV/m (Fig. 1), and our $\pm E_c$ and $\pm P_r$ values for bulk and pre-stressed actuators were higher than those obtained by Dausch, probably due to the higher electric fields (PZT 5H: $E_{c\text{bulk}} = 0.36$ MV/m, $E_{c\text{RAINBOW}} = 0.42$ MV/m and PZT 5A: $E_{c\text{bulk}} = 0.65$ MV/m, $E_{c\text{RAINBOW}} = 0.84$ MV/m and) [3]. PZT 5A THUNDER actuators had higher remnant polarisation and about the same coercive electric field under ~2 MV/m ($+E_c$ = 1.38 MV/m and $-E_c = -1.4$ MV/m) than pre-stressed actuators presented here. At higher electric fields our remnant polarisation values were closer to the values of THUNDER actuators and the coercive electric field was higher. However, it should be noted that the thickness of the THUNDER actuators was ∼200 μm which has effect on properties as later introduced [3, 11]. Similarly, passive ring has hindering effect for polarisation as stated earlier.

Typically, the $\pm E_c$ values were greater with thinner components (Table 1). Thickness had an effect on the P_r values when the sample was pre-stressed, whereas within the bulk samples, thickness had no high significance. In other words, thicker pre-stressed actuators had P_r values closer to those of the bulk counterparts than did the thinner ones. This result indicates correlation between the pre-stress level and dielectric properties of the actuators as reported in many papers for different structures. Since the same pre-stressing element was used in every actuator only thickness of the PZT material and its material properties define the difference in pre-stress state of presented actuators.

The results also showed that the bulk actuators and pre-stressed 250 μm thick PZT 5H actuator had slightly higher $-P_r$ than $+P_r$ values and the behaviour was opposite that of the rest of the pre-stressed actuators (250 μm and 375 μm thick PZT 5A and 375 μm thick PZT 5H, Table 1). In addition, these same actuators had higher $+Ec$ values and actuators having higher $+Pr$ had higher $-Ec$. Partially same results was obtained by Ounaies et al. with PZT 5A THUNDER actuators (e.g. $+E_c = 10$ MV/m, $-E_c = -15.5$ MV/m, $+P_r = 34$ μC/cm^2 and $-P_r = -32.5\mu$C/cm^2) [15]. Behaviour is due mechanically biased stress state, favourable to switch domains at certain direction. Stress-induced-domain reorientation is described more detailed by Haertling [20]. Actuators presented here are very similar with RAINBOW actuators in the sense that pre-stressing layer (fired metal and lead, respectively) in both cases are not over the whole area of the actuators leaving passive ring area. In the case of the RAINBOW actuators the ring is often made active during electrode manufacturing that can cause some differences in results.

In general, the differences between these results and those obtained by others can be explained by the differences in pre-stress level, stronger clamping of the thicker passive layers of the RAINBOW and THUNDER actuators and passive ring area introducing high tensile stresses [3, 11]. In thin films pre-stressing can alter the dielectric constant, remnant polarisation and coercive electric field of the actuators through tensile or compressive stress. The behaviour of thin film actuators depends on composition as well as the amount of pre-stressing through the thickness of the samples [21, 22]. A similar effect has also been reported by Zhao et al. for bulk samples with uniaxial compressive stress, e.g., PZT 5H material [28, 29]. The behaviours presented here are caused by the intrinsic transverse tensile and compressive stresses of the actuators introduced by pre-stressing layer and passive insulation area. Profound stress analysis of the actuators is needed for further investigation. In terms of obtaining high displacement, proper level of tensile stresses can increase the effective d_{31} coefficient utilised in benders [10, 12, 23, 24]. Although the actuators manufactured here had nearly the same curvature as the RAINBOW counterparts, they had a relatively thin metal layer after firing (∼30 μm) to provide pre-stress and clamping of the bottom layer. This pre-stressing layer is much thinner than the passive layers in THUNDER and RAINBOW actuators causing difference in e.g. constraining of the piezo displacement which creates bending actuation.

Figures 6 and 7 show the typical normalised displacement, dielectric constant and positive remnant polarisation values of the pre-stressed PZT 5H actuators (thickness 375 and 250 μm) poled at 25 and 125°C temperatures.

These results show that the different poling fields had different reductions in displacement and dielectric constant due to ageing. A clear reason for this could not be found within this study, but fatigue characteristics will be investigated profoundly in the future. The piezoelectric coefficient d_{33}, d_{31} and remnant polarisation have a clear connection in the case of the bulk

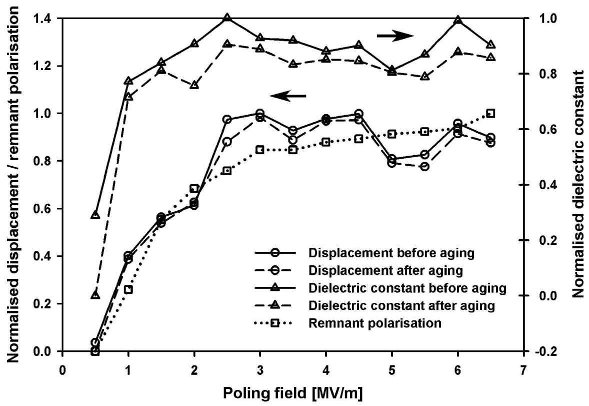


Fig. 6. Normalised displacement, remnant polarisation and dielectric constant of the pre-stressed PZT 5H actuator (thickness 375 μm) as a function of poling field (at 25°C) before and after aging.

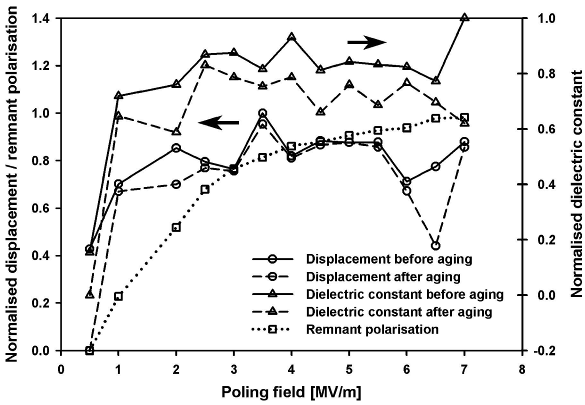


Fig. 7. Normalised displacement, remnant polarisation and dielectric constant of the pre-stressed PZT 5H actuator (thickness 250 μm) as a function of poling field (at 125°C) before and after aging.

actuators [6, 23, 25]. Similar relation was found here between displacement and remnant polarization. However, in this case d_{31} is an effective piezoelectric coefficient which is transferred to axial displacement through large bending deformation that can have also other affecting parameters. The remnant polarisation curve in Fig. 6 correlates well with the displacement curve after poling, but more comparable results was obtained with a dielectric constant measured with a small signal. This is probably because the small signal measured actual state of ceramic after poling, without enforcing high electric fields that had effect on properties. In Fig. 7 remnant polarisation is not as comparable with displacement as in the case of Fig. 6. The reason for this is not known, but it still gives a decent estimation of the displacement of the component. Neither was the dielectric constant as accurate as in Fig. 6, and the most probable cause is that the small signal measurements were done at room temperature, while poling was carried out at 125°C. In both cases (Figs. 6 and 7), displacement is saturated much before the maximum electric field, and therefore more displacement is not achieved using higher poling fields. However, as the earlier figures show (Figs. 1, 3, and 5), the coercive electric field increases at higher poling fields, justifying their use for poling which ultimately increases the displacements through higher operating electric fields. Correlation of remnant polarisation and permittivity with displacement as a function of poling temperature needs further investigations.

The measurements covered a total of 60 samples with small variation as specified by the manufacturer. Regardless of the manufacturer's tolerances, the behaviours of the actuators were the same as presented here.

Thus, the main result of this research is that characterisation of the hysteresis loop (remnant polarization and coercive electric field) is a useful tool also when optimising the poling of pre-stressed piezoelectric bender actuators. This is true even though the displacement of bending actuators is a function of their length and thickness, and different materials have different piezoelectric coefficients resulting in a different ratio between d_{31} and P_r [5, 26].

4. Conclusions

The results show that measurement of the hysteresis curve values can be used for prediction and optimisation of the discrete poling conditions of the new pre-stressed piezoelectric bender actuators realised with post-fired biasing layer. This is an important result, since it was shown that poling of a pre-stressed bender actuator must be done more carefully than that of bulk counterparts. The remnant polarisation and coercive electric field values of a pre-stressed actuator depend more gradually on the poling field than in the case of the bulk counterpart. The higher remnant polarisation and coercive field values were mainly available at the lowest studied temperature (25°C). The remnant polarisation and coercive electric field results with different materials and component thicknesses also support the conclusion that they are largely changed by the pre-stress state inside the ceramic. According to modelling results and measurements large tensile stresses occurs due passive insulation area which hinders polarization of the bulk actuators. Same behaviour but lower stresses are expected to occur with pre-stressed bending actuators, due smaller in-plane displacement. Finally, the results show that remnant polarisation gave a decent estimation of displacement as a function of the poling field, and so it can be used in the optimisation of the poling of pre-stressed actuators, in spite of the fact that the actuators experienced large deformations and stresses in poling and actual use. Additionally, small signal measurement of the dielectric constant gave even better correlation with displacement, probably since it does not induce as high electric field as the poling effect itself, and it can be used for quick final checking of actuator performance after poling.

Acknowledgments

This work was completed within the "Novel ceramic technologies in realisation of miniature actuators and motors" project funded by the Academy of Finland (project 49830, TUKEVA programme) and the Finnish National Technology Agency (project 52059, PRESTO programme), as well as the "Multifunctional ceramic packages" project funded by the Academy of Finland (project 206123). Jari Juuti also acknowledges the financial support of the Tauno Tönning Foundation. The authors also wish to thank Pasi Leppäniemi for his help with the measurements.

References

1. K. Bhattacharya and G. Ravichandran, *Acta Materialia*, **51**, 5941 (2003).
2. D.H. Pearce and T.W. Button, in *Proceedings of the Eleventh IEEE International Symposium on Applications of Ferroelectrics*, (IEEE, Piscataway, 1998), p. 547.
3. D.E. Dausch, *Journal of American Ceramic Society*, **80**(9), 2355 (1997).
4. Q. Jiang, W. Cao, and L.E. Cross, in *Proceedings of the Eighth International Symposium on Applications of Ferroelectrics*, (IEEE, New York, 1992), p. 107.
5. W.Y. Shih, W.-H. Shih, and I.A. Aksay, *Journal of American Ceramic Society*, **80**(5), 1073 (1997).
6. H. Wang, R.E. Newnham, L.E. Cross, and W.Y. Pan, in *IEEE Seventh International Symposium on Applications of Ferroelectrics* (IEEE, New York, 1990), p. 422.
7. B.W. Barron, G. Li, and G.H. Haertling, in *Proceedings of the Tenth IEEE International Symposium on Applications of Ferroelectrics* (IEEE, New York, 1996), p. 305.
8. X. Li, J.S. Vartuli, D.L. Milius, I.A. Aksay, W.Y. Shih, and W.-H. Shih, *Journal of American Ceramic Society*, **84**(5), 996 (2001).
9. S.A. Wise, *Sensors and Actuators*, **A 69**, 33 (1998).
10. X. Li, W.Y. Shih, J.S. Vartuli, D.L. Milius, I.A. Aksay, and W.-H. Shih, *Journal of American Ceramic Society*, **85**(4), 844 (2002).
11. Z. Ounaies, K. Mossi, R. Smith, and J. Bernd, *ICASE report*, No. 2001-9, NASA/CR-2001-210859, April 2001.
12. R.W. Schwartz, L.E. Cross, and Q.M. Wang, *Journal of American Ceramic Society*, **84**(11), 2563 (2001).
13. J. Juuti, E. Heinonen, V.-P. Moilanen, and S. Leppävuori, *Journal of the European Ceramic Society*, **24**, 1901 (2004).
14. K.M. Mossi, G.V. Selby, and R.G. Bryant, *Materials Letters*, **35**, 39 (1998).
15. J. Mulling, T. Usher, B. Dessent, J. Palmer, P. Franzon, E. Grant, and A. Kingon, *Sensors and Actuators*, **A 94**, 19 (2001).
16. R.W. Schwartz and M. Narayanan, *Sensors and Actuators*, **A 101**, 322 (2002).
17. H.-W. Wang, S.-Y. Cheng, and C.-M. Wang, in *Proceedings 1989 Japan International Electronic Manufacturing Technology Symposium* (IEEE, New York, 1989), p. 263.
18. J. Juuti, H. Jantunen, V.-P. Moilanen, and S. Leppävuori, *submitted to IEEE Transactions on Ultrasonics, Ferroelectrics and Frequency Control*.
19. H. Moilanen and S. Leppävuori, *Sensors and Actuators*, **A 92**, 236 (2001).
20. G.H. Haertling, in *Proceedings of the Tenth IEEE International Symposium on Applications of Ferroelectrics* (IEEE, Piscataway, 1996), p. 65.
21. W.E. Paradise, F. Wang, and G.H. Haertling, in *Proceedings of the Ninth IEEE International Symposium on Applications of Ferroelectrics* (IEEE, New York, 1994) p. 784.
22. J.F. Jr. Shepard, S. Trolier-McKinstry, M.A. Hendrickson, and R. Zeto, in *Proceedings of the Tenth IEEE International Symposium on Applications of Ferroelectrics* (IEEE, New York, 1996), p. 161.
23. J. Zhao and Q.M. Zhang, in *Proceedings of the Tenth IEEE International Symposium on Applications of Ferroelectrics* (IEEE, New York, 1996), p. 971.
24. J. Zhao, Q.M. Zhang, and V. Mueller, in *Proceedings of the Eleventh IEEE International Symposium on Applications of Ferroelectrics* (IEEE, Piscataway, 1998), p. 361.
25. Q.M. Zhang, J. Chen, and L.E. Cross, in *IEEE 1993 Ultrasonics Symposium Proceedings* (IEEE, New York, 1993), p. 525.
26. V.D. Kugel, S. Chandran, and L.E. Cross, in *Proceedings of the SPIE The International Society for Optical Engineering* (SPIE—Int. Soc. Opt. Eng., 1997), p. 70.
27. M. Sayer, B.A. Judd, K. EI-Assal, and E. Prasad, *Journal of Canadian Ceramic Society*, **50** (1981).
28. T. Tanimoto and K. Okazaki, in *Proceedings of the Eighth IEEE International Symposium on Applications of Ferroelectrics* (IEEE, New York, 1992), p. 504.
29. Q.-M. Wang and L.E. Cross, *Journal of American Ceramic Society*, **82**(1), 103 (1999).