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Degradation of the d_{33} piezoelectric coefficient for PZT ceramics under static and cyclic compressive loading

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Abstract

The degradation of the d_{33} piezoelectric coefficient of PZT ceramics subjected to compressive stresses along the poling direction was studied in the range from 10 to 70 MPa in static and cyclic loading. The coefficient was measured making use of the direct piezoelectric effect with the same servohydraulic test machine used to apply the stresses. The piezoelectric coefficient was measured as a function of the amplitude of the applied stress, which allowed us to isolate the intrinsic coefficient and the domain wall contribution. A hard and a soft piezoelectric ceramic were studied. The hard piezoelectric material was very resistant to degradation in the range of stress investigated. The soft material showed significant piezoelectric degradation due to stress induced depolarisation. The degradation was significantly higher for a given stress in cyclic loading than in static loading. © 2001 Elsevier Science Ltd. All rights reserved.

Keywords: Degradation; Piezoelectric properties; PZT

1. Introduction

Piezoelectric ceramics are usually preloaded with a compressive stress in actuation applications because they are easily fractured under tensile stresses.[1] Many of these applications involve high frequency and relatively high amplitude driving electric fields.[2] Therefore, the ceramics are cyclically tensile strained (by the piezoelectric effect) during operation under the compressive static load, which introduces an additional high frequency component in the compressive stress. The total compressive stress must be kept below the values at which depolarisation and microcracking occur.[3] These two effects lead to the degradation of the piezoelectric coefficients.[4]

We present here a study of the degradation of the d_{33} piezoelectric coefficient in static and cyclic compressive loading along the poling direction for two, one hard and one soft, commercial lead zirconate titanate (PZT) piezoelectric ceramics. The piezoelectric coefficient was measured before and after the mechanical treatments by the direct measurement of the piezoelectric charge generated during the application of an uniaxial stress sine wave. This measurement was accomplished in the same servohydraulic test machine used to apply the treatments, and allowed us to vary the amplitude of the stress sine wave. This enabled us to study piezoelectric nonlinearities.[5] The coefficients obtained were compared with those provided by the more standard resonance technique. A significant degradation of d_{33} was observed for the soft ceramic for stresses as low as 10 MPa.

2. Experimental procedures

The piezoelectric ceramics were two $(Pb,Sr)(Zr,Ti)O_3$ based compositions, referred to as PZT-4D and PZT-5H, which included minor dopants that gave them their hard and soft character, respectively. Details of their properties can be found in the supplier's catalogue.[6] Ceramics were received as poled plates with Ag sintered electrodes, from which specimens with dimensions suitable for the piezoelectric characterisation were cut. The dimensions of the original plates did not allow us to make use of the existing polarisation. Therefore, the Ag electrodes were removed by polishing with diamond paste and the specimens depoled with a thermal treatment above the

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Curie temperature. New Au electrodes were deposited by sputtering on the appropriate faces and the samples repoled at 2.5 kV mm^{-1} and 100°C.

The longitudinal piezoelectric coefficient, d_{33}, was measured by the direct measurement of the piezoelectric charge generated during the application of an uniaxial stress sine wave. $10\times5\times5$ mm^3 specimens poled along the 10 mm direction were prepared for the two compositions. Measurements were performed in a servohydraulic test machine (Instron 8500), to which a charge to voltage converter (105 nC V^{-1}) had been incorporated. This measured the electric charge generated by the application of stress. Stress sine waves of 0.5, 1, 1.5, 2 and 2.5 MPa amplitude, σ_{ac}, and of 1 Hz frequency were applied to the sample, which had been pre-loaded with a compressive stress of 5 MPa (see Fig. 1(a)). The piezoelectric coefficient was evaluated as the ratio between the charge generated and the force applied as a function of σ_{ac}. The results of d_{33} vs. σ_{ac} were fitted to the expression (Eq. (1)):

$$d_{33} = d_{33}^{int} + \alpha\sigma_{ac} \quad (1)$$

where d_{33}^{int} is the intrinsic (without domain wall contributions) piezoelectric coefficient[7] and α a measurement of piezoelectric non-linearities related to domain wall contributions.[8]

The d_{33} coefficient was measured before and after compressive loading along the poling direction for stresses ranging from 10 to 70 MPa. Two different loading profiles were applied:

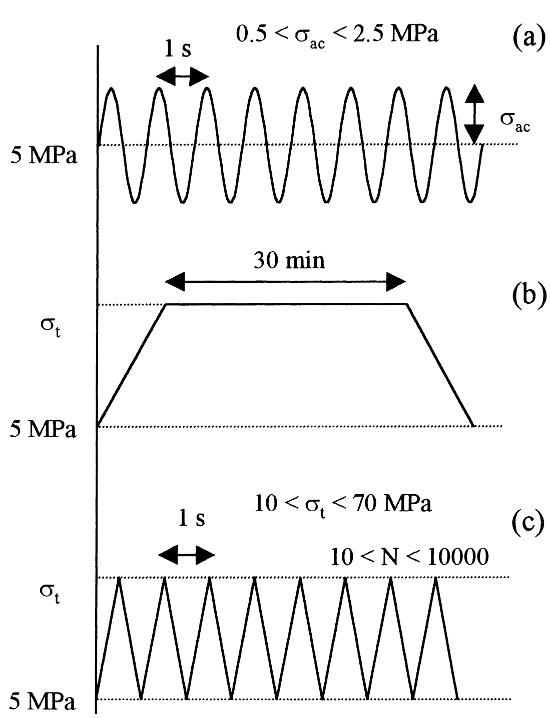

Fig. 1. Stress profiles: (a) for the d_{33} measurement; (b) for degradation in static loading; and (c) for degradation in cyclic loading.

- From 5 MPa to the loading stress, σ_t, at 4 MPa min^{-1}, 30 min at σ_t, and back to 5 MPa at −4 MPa min^{-1} — static loading [see Fig. 1(b)].
- A train of triangular pulses from 5 MPa to σ_t with a 1 Hz frequency and increasing (from 10 to 10, 000) number of pulses — cyclic loading [see Fig. 1(c)].

The d_{33} coefficient for the PZT-5H ceramics was also measured before mechanical loading by the resonance technique.[9] $10\times3\times3$ mm^3 bars poled along the 10 mm direction were prepared for this measurement. These measurements are performed at high frequencies (the resonance frequencies are typically tens of kHz) compared to the 1 Hz used for the measurements with the test machine.

3. Results and discussion

An example of the results of d_{33} vs. σ_{ac} and of their fit to Eq. (1) is given in Fig. 2 for the two compositions. The obtained piezoelectric parameters: d_{33}^{int} and α/d_{33}^{int} at 1 Hz for the two type of ceramics, and d_{33} at 127 kHz for the PZT-5H ceramics (from the resonance method), after poling, prior to degradation testing, are given in Table 1. α/d_{33}^{int} is given because the ratio gives a more representative measure of the level of piezoelectric non-linearity relative to their d_{33}^{int} values. The level of Sr substituting for Pb in the A site of the perovskite was higher for the PZT-5H material than for the PZT-4D, which gives the former composition a lower Curie temperature,[6] and thus, a higher intrinsic coefficient. The α/d_{33}^{int} ratio was also higher for the PZT-5H composition as a consequence of its softer character, because of a higher mobility of the ferroelastic domain walls. There

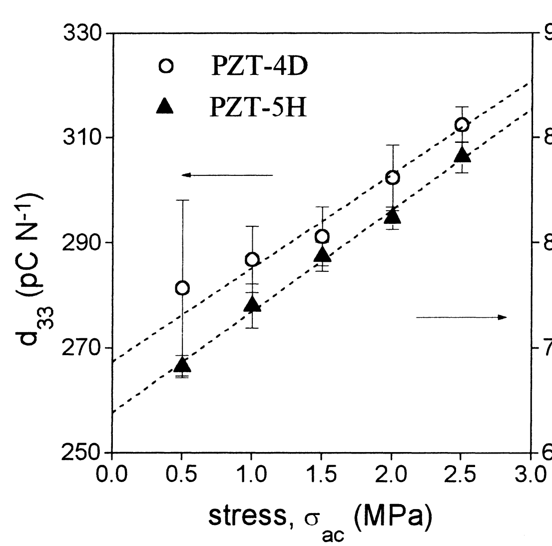

Fig. 2. d_{33} vs. σ_{ac} after poling for the two compositions.

Table 1
Piezolectric parameters of the piezoceramics after poling prior to mechanical loading

	$d_{33}^{\rm int}$ at 1 Hz $\times 10^{-12}$ C N^{-1}	$\alpha/d_{33}^{\rm int}$ at 1 Hz $\times 10^{-6}$ m^2 N^{-1}	d_{33} at 127 kHz $\times 10^{-12}$ C N^{-1}
PZT-4D	246±27	0.08±0.05	–
PZT-5H	677±18	0.11±0.03	689±15

is good agreement between the d_{33} provided by the resonance technique and $d_{33}^{\rm int}$.

The changes of both $d_{33}^{\rm int}$ and $\alpha/d_{33}^{\rm int}$ after static loading at increasing stress, expressed as % of the initial values (see Table 1), are shown in Fig. 3. We were aware that depolarisation under static loading does not occur instantaneously but can have a time dependence, which is a consequence of the slow movement of the ferroelastic domain walls under the stress, which manifests itself as mechanical creep.[10] Our system allowed us to monitor the charge transient associated with this creep under static loading, and it was found to essentially arrest ($<7\times10^{-6}$ C m^{-2} s^{-1}) after 10 to 20 min. 30 min was then chosen as a sufficient time to have exhausted this process. The intrinsic coefficient for the PZT-4D piezoceramics was hardly affected by static compressive loading in the stress range investigated (see Fig. 3(a)), indicating the good resistance of this composition against stress induced depolarisation. In comparison, the intrinsic coefficient for the PZT-5H piezoceramics showed a strong degradation. This degradation was fully reversible by repoling, indicating that it was caused by stress induced depolarisation. The coefficient dropped to 50% of its initial value after loading at 30 MPa, and dropped to 25% after loading at 70 MPa. The occurrence of depolarisation for the PZT-5H material and lack of it for the PZT-4D, was confirmed by measuring the electrical charge generated during the application of a compression loop to 70 MPa. The results are shown in Fig. 4. Note that all the charge generated during loading for the PZT-4D ceramic was fully reversed during unloading, but it was not for the PZT-5H sample. $\alpha/d_{33}^{\rm int}$ for the PZT-4D ceramics increased to a 138±11%of its initial value after loading, but a systematic trend with the loading stress could not be stated. This increase in the ferroelastic domain wall mobility might be related to a deageing effect. In acceptor doped piezoceramics (hard ceramics), the ferroelastic domain wall mobility is known to be limited by the presence of dopant-oxygen vacancy complexes, which have a dipolar moment that aligns with the spontanous polarisation within a domain.[11] The deageing would consist of the partial disalignment of the complexes induced by the stress. $\alpha/d_{33}^{\rm int}$ for the PZT-5H is hardly affected. This indicates that there is not a significant change in the environment of the domain walls after applying the stress.

The changes of $d_{33}^{\rm int}$ after cyclic loading at increasing number of pulses, N, expressed as % of the initial values (see Table 1), are shown in Fig. 5 for different stress values, σ_t. The changes were fully reversible by repoling. The levels of degradation in static loading are included in the figure as dotted lines for comparison. The intrinsic

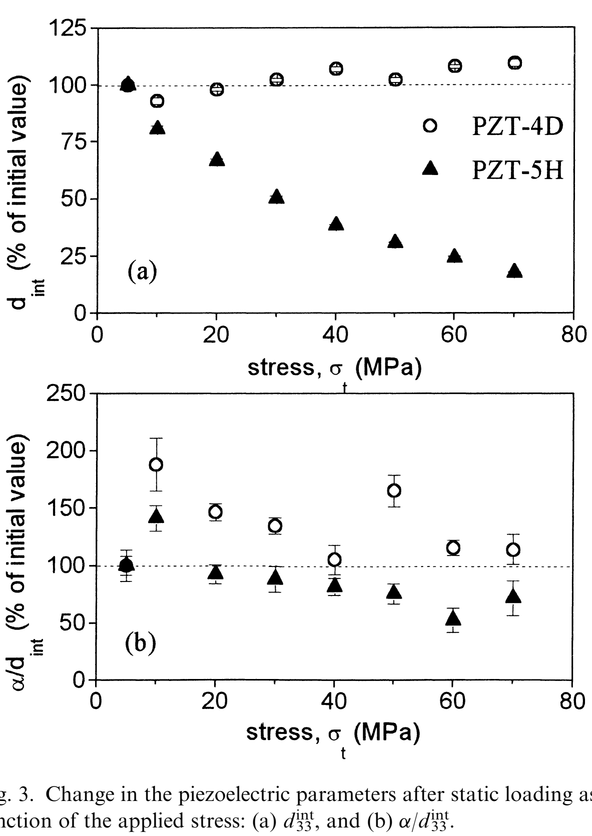

Fig. 3. Change in the piezoelectric parameters after static loading as a function of the applied stress: (a) $d_{33}^{\rm int}$, and (b) $\alpha/d_{33}^{\rm int}$.

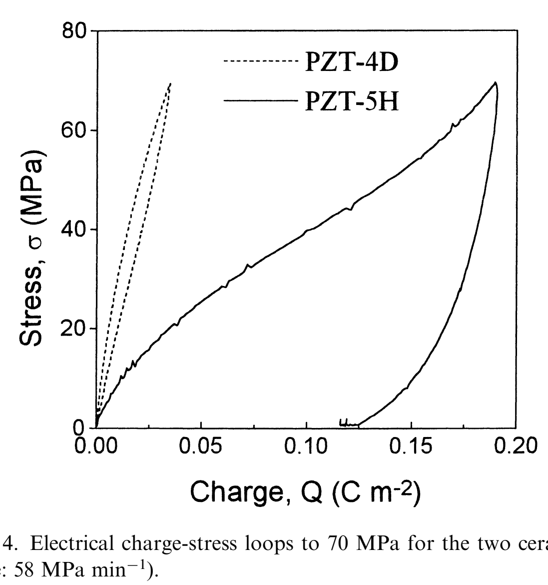

Fig. 4. Electrical charge-stress loops to 70 MPa for the two ceramics (rate: 58 MPa min^{-1}).

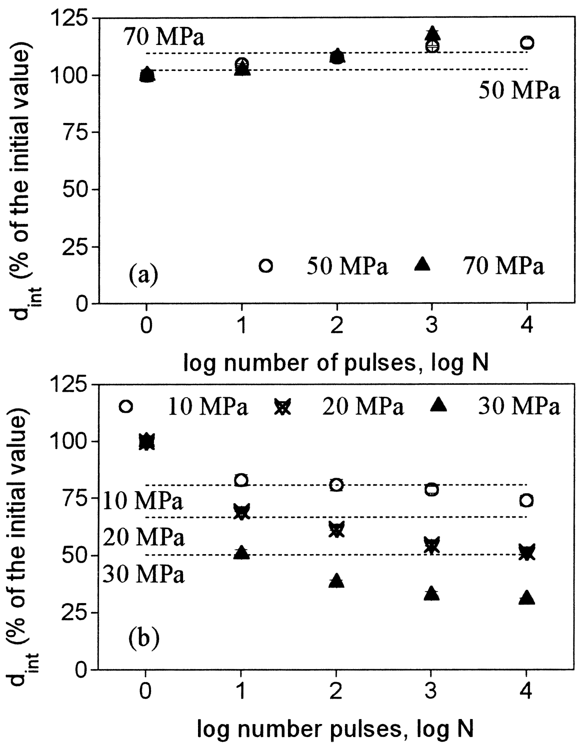

Fig. 5. Change in d_{33}^{int} after cyclic loading as a function of the number of pulses for different applied stress: (a) PZT-4D; and (b) PZT-5H. The dashed lines correspond to the values for static loading.

coefficient for the PZT-4D piezoceramics was not significantly afftected by cyclic compressive loading. The point corresponding to 10 000 pulses at 70 MPa is missing because the sample fractured along the poling direction during cycling. The intrinsic coefficient for the PZT-5H piezoceramics showed a significant additional degradation in cyclic loading compared to a single static loading. This degradation was already higher after 1800 pulses than that achieved after 30 min under static loading at the maximum cyclic stress, and continued to increase with further cycling. Recall that depolarisation in static loading was exhausted after 30 min.

α/d_{33}^{int} for the PZT-4D ceramics decreased after cyclic loading, contrary to what it did in static loading. This seems a surprising result for which we can offer no explanation. More research is in progress to investigate this topic. α/d_{33}^{int} for the PZT-5H ceramics was not significantly affected by cyclic loading consistent with the static loading results.

4. Conclusions

The intrinsic piezoelectric coefficient, d_{33}^{int}, for PZT-4D hard piezoceramics was highly resistant to degradation under static and cyclic compressive loading along the poling direction in the range 10–70 MPa, indicating the absence of stress induced depolarisation. The piezoelectric non-linearities slightly increased after static loading. This increase of the ferroelastic domain wall mobility was most probably linked to a deageing effect. The d_{33}^{int} for PZT-5H soft piezoceamics strongly degraded after compressive loading, to 50% of its value after poling after static loading at 30 MPa, and to 25% after loading at 70 MPa. The level of non-linearity is not affected by stress for the soft ceramic. The degradation in d_{33}^{int}, and so in d_{33}, is significantly higher in cyclic loading than in static loading for a given stress. Therefore, the thresholds for depolarisation evaluated by static loading are meaningless for applications involving cyclic stresses, and that a specific characterisation, as done here, is necessary to predict long-term behaviour.

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