

HIGHLY ANISOTROPIC
ELECTROMECHANICAL PROPERTIES
IN MODIFIED LEAD-TITANATE
CERAMICS

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Highly anisotropic electromechanical properties in modified lead titanate ceramics

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The Pennsylvania State University, 1987



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Highly Anisotropic
Electromechanical Properties
in Modified Lead Titanate Ceramics

A Thesis in
Ceramic Science

by

Dragan M. Damjanovic

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Dragan M. Damjanovic
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ABSTRACT

The dielectric, elastic and piezoelectric properties of modified lead titanate ceramics were investigated with the goal of better understanding their highly anisotropic electromechanical behavior.

Two compositions, $(\text{Pb}_{0.85}\text{Sm}_{0.10})(\text{Ti}_{0.98}\text{Mn}_{0.02})\text{O}_3$ (or PST) and $(\text{Pb}_{0.76}\text{Ca}_{0.24})[(\text{Co}_{1/2}\text{W}_{1/2})_{0.04}\text{Ti}_{0.96}]\text{O}_3$ (or PCT), were investigated as representatives of modified lead titanate ceramics. Measurements of dielectric, elastic and piezoelectric properties of PST and PCT ceramics, using a standard resonance technique, showed that the large anisotropy in the electromechanical coupling factors is exclusively due to the anisotropy in the piezoelectric coefficients d_{33} and d_{31} . The ratio d_{33}/d_{31} depends on both poling field and temperature.

The thickness coupling factor, k_t , remains almost constant with temperature for PCT and PST ceramics. At room temperature d_{33} increases with poling field, saturating to near 60 pC/N at large poling fields. At the same time d_{31} initially increases and then decreases with poling field thus causing a large d_{33}/d_{31} ratio.

Measurements of complex material coefficients relevant for the planar coupling mode, using an iterative technique, showed that d_{31} as well as the elastic compliance, s_{11}^E , and the dielectric permittivity, ϵ_{33}^X , must be considered as complex in order to accurately describe the resonant properties of these ceramics. Measuring the complex d_{31} as a function of temperature (-180°C to +100°C), it was possible to show that the real component d_{31}' , in both ceramics, changes sign with temperature, thus causing a zero planar coupling factor, k_p , at a certain temperature. The examination of the dependence of complex d_{31} on temperature with poling field as a parameter (10 to 90 kV/cm) showed that the temperature at which d_{31}' becomes zero depends on the poling field. This shift of zero crossover point of d_{31}' with poling field causes a decrease of d_{31}' (and k_p) with poling field at room temperature.

A significant imaginary component of the piezoelectric coefficient and a similar temperature behavior of imaginary elastic, dielectric and piezoelectric coefficients in these ceramics indicate a presence of piezoelectric relaxational mechanisms, whose origin is in a coupling between elastic and dielectric losses. Assuming that the complex d_{31} in PST and PCT ceramics may be represented as a sum of the intrinsic piezoelectric effect, d_{31}^{∞} , and extrinsic relaxational contributions, $\Delta d_{31}' - id_{31}''$, it is possible to consistently interpret the dependence of the experimentally determined real part, $d_{31}' = d_{31}^{\infty} + \Delta d_{31}'$, on poling field and temperature through competing effects of d_{31}^{∞} and $\Delta d_{31}'$.

A large anisotropy in piezoelectric coefficients seems to be only enhanced by zero d_{31} since a large d_{33}/d_{31} ratio was observed even in those samples which, due to differing processing conditions, did not exhibit a change of sign in d_{31}' over the investigated temperature range. This large anisotropy in piezoelectric coefficients seems to be a consequence of averaging of single crystal properties of modified lead titanate ceramics.

The neutron powder diffraction study showed that Sm in PST ceramics was properly incorporated at the A site in the perovskite structure. A model with Ca cation substituting only for Pb as well as a model with Ca randomly occupying the A and B site lead to unrealistic stoichiometries of PCT composition. In both compositions as well as in a pure PbTiO_3 powder, the shifts of O1 and O2 oxygens in respect to their ideal cubic positions are different, causing an asymmetric oxygen octahedron.

The equivalent circuit of a piezoelectric resonator with a large piezoelectric phase angle was derived allowing that it may include a frequency dependent resistive term. This was necessary in order to describe piezoelectric contributions to the total energy dissipation of the resonator.

TABLE OF CONTENTS

	Page
ABSTRACT	iii
LIST OF TABLES	ix
LIST OF FIGURES	x
ACKNOWLEDGEMENTS	xvi
Chapter	
1 INTRODUCTION.	1
1.1 Introductory remarks on piezoelectric materials and piezoelectricity	1
1.2 Outline of the thesis.	9
1.3 Basic piezoelectric relations	10
1.3.1 Stress and strain	10
1.3.2 Electric field and polarization	11
1.3.3 Coupling between the elastic and dielectric properties	12
1.3.3.1 Piezoelectric effect	12
1.3.3.2 Electrostrictive effect	16
1.3.4 Symmetry considerations	17
1.3.4.1 Matrix notation	17
1.3.4.2 Thermodynamic arguments	18
1.3.4.3 Neumann's principle	19
1.3.5 Vibrations of piezoelectric body.	20
1.3.5.1 Introduction	20
1.3.5.2 Lengthwise vibrations of a bar with electric field perpendicular to length	23
1.3.5.3 Lengthwise vibrations of a bar with electric field parallel to length	27
1.3.5.4 Radial vibrations of a thin piezoelectric disk	28
1.3.5.5 Thickness vibrational modes	29

TABLE OF CONTENTS (cont.)

Chapter	Page
1.3.6 Electromechanical coupling factors	32
1.3.7 Equivalent circuit	36
1.3.7.1 Introduction	36
1.3.7.2 Equivalent circuit of a lossy resonator	38
1.4 Complex piezoelectric coefficients	44
1.4.1 Introduction	44
1.4.2 Dielectric and elastic relaxation	45
1.4.3 Piezoelectric relaxation	53
1.4.3.1 Effects of symmetry on relaxation of single crystals	58
1.4.3.2 Complex piezoelectric coefficients and energy dissipation	60
1.4.4 Experimental studies of relaxation in piezoelectric coefficients	64
1.4.4.1 90° domain wall contribution to the piezoelectric coefficients in ferroelectric ceramics	65
1.4.4.2 Piezoelectric relaxation in single crystal AgNa(NO ₂) ₂	70
1.4.4.3 Piezoelectric relaxation in PZT-epoxy composite system	74
2 ELECTROMECHANICAL AND RELATED PROPERTIES OF FERROELECTRIC MATERIALS BASED ON LEAD TITANATE	78
2.1 Properties of single crystals of lead titanate	78
2.1.1 Crystal structure of lead titanate.	78
2.1.2 Dielectric, elastic, and piezoelectric properties of single crystals of lead titanate	85
2.2 Properties of the lead titanate ceramics with small amounts of additives	88
2.3 Lead titanate ceramics modified with alkaline and rare earths.	97
2.3.1 Lead titanate ceramics modified with alkaline earths	97
2.3.2 Lead titanate ceramics modified with rare earths	104
2.4 Other systems with a large electromechanical anisotropy.	114

TABLE OF CONTENTS (cont.)

Chapter	Page
3 STATEMENT OF THE PROBLEM	115
4 MATERIAL PREPARATION AND MEASUREMENTS	118
4.1 Selection of materials	118
4.2 Material preparation	118
4.2.1 Preparation of $(\text{Pb}_{0.85}\text{Sm}_{0.10})(\text{Ti}_{0.98}\text{Mn}_{0.02})\text{O}_3$ ceramics.	118
4.2.2 Preparation of $(\text{Pb}_{0.74}\text{Ca}_{0.26})[(\text{Co}_{1/2}\text{W}_{1/2})\text{Ti}_{0.96}]\text{O}_3$ ceramics	120
4.2.3 Cutting and polishing of the samples	122
4.2.4 Electroding and poling	124
4.3 Dielectric measurements	126
4.4 Piezoelectric measurements	127
4.4.1 Planar coupling factor	127
4.4.1.1 Calculations of Poisson's ratio and related coefficients from radial vibrations of a disk.	128
4.4.1.2 Measurements of the resonant frequencies f_s and f_p for the radial mode resonance	131
4.4.2 Measurements of complex material coefficients of a length extensional bar with electric field perpendicular to its length	133
4.4.2.1 Calculation of complex coefficients	134
4.4.2.2 Measurements of $Y = G + iB$ for a resonating bar	138
4.4.3 Thickness coupling factor	139
4.4.4 Quasistatic d_{33} coefficient	140
5 RESULTS AND DISCUSSION	141
5.1 Dielectric permittivity of PCT and PST ceramics at subresonant frequencies	141
5.2 Planar coupling factor as a function of poling field and temperature	145
5.2.1 Dependence on poling field.	145
5.2.2 Dependence on temperature.	154
5.2.3 Summary	168

TABLE OF CONTENTS (cont.)

Chapter	Page
5.3 Thickness coupling factor as a function of poling field and temperature	168
5.3.1 Dependence on poling field.	168
5.3.2 Dependence on temperature.	171
5.3.3 Summary	177
5.4 Complex dielectric, elastic and piezoelectric coefficients	177
5.4.1 An evidence of a nonzero piezoelectric phase angle in modified lead titanate ceramics	178
5.4.2 Sign of d_{31} in PST and PCT ceramics.	182
5.4.3 Dependence of complex d , s and ϵ on temperature and poling field	189
5.4.4 Summary	195
5.5 Possible mechanisms for the highly anisotropic electromechanical properties in modified lead titanate ceramics	198
5.5.1 Polycrystallinity and apparent piezoelectric coefficients in modified lead titanate ceramics	198
5.5.2 Relaxational contributions to piezoelectric coefficients	203
5.5.3 Microstructural considerations	218
5.5.4 Low temperature anomalies	224
5.5.5 Crystal structure of PST and PCT ceramics	230
5.6 A comment on equivalent circuit of a resonator with a large piezoelectric phase angle	234
6 SUMMARY AND RECOMMENDATIONS FOR FUTURE WORK	241
6.1 Summary	241
6.2 Suggestions for future work.	244
BIBLIOGRAPHY	247

LIST OF TABLES

Table		Page
1	Matrices of the elastic compliances s , dielectric permittivity ϵ and piezoelectric coefficient d for selected point groups	21
2	Dielectric, elastic and piezoelectric coefficients of selected piezoelectric materials	35
3	Values of dielectric, piezoelectric, elastic and electrostrictive coefficients of a PbTiO_3 single crystal at room temperature.	86
4	Summary of Rietveld analysis of modified lead titanate ceramics	231

LIST OF FIGURES

Figure		Page
1	Geometries of common piezoelectric resonators	24
2	Admittance of a piezoelectric resonator	26
3	Admittance of a PZT piezoelectric disk near the fundamental resonant frequency of the thickness mode resonance	31
4	Equivalent circuit of a piezoelectric resonator near resonance	37
5	(a) Susceptance, B, and conductance, G, and (b) resistance, R, and reactance, X, of a piezoelectric resonator near an isolated resonance	41
6	(a) Admittance and (b) impedance loops of a piezoelectric resonator near an isolated resonance	42
7	Schematic illustration of relaxation behavior	47
8	The real and imaginary parts of a complex material coefficient m as a function of frequency according to the Debye equations (73) and (74)	51
9	A two-dimensional model of a material with defects that are simultaneously elastic and electric dipole	54
10	The potential energy of a defect in the bistable model	56
11	90° domains in tetragonal ceramics	66
12	Measured and extrapolated real and imaginary parts of the dielectric constant, the elastic compliance and the piezoelectric constant as a function of temperature in Fe-doped PZT ceramics	69
13	Cole-Cole plots of the complex piezoelectric constants in $\text{AgNa}(\text{NO}_2)_2$	71
14	Plots of d' and d'' in the complex plane for Epoxy-PZT composite	76
15	Perovskite structure	79
16	Temperature dependence of k_{31} and d_{31} for a single crystal of PbTiO_3	87

LIST OF FIGURES (cont.)

Figure		Page
17	(a) Coupling factors k_t and k_p and (b) temperature coefficient N_p of frequency constant of radial mode vs. amount of alkaline earth metals in $(\text{Pb}_{1-x}\text{Me}_x)[(\text{Co}_{1/2}\text{W}_{1/2})_{0.04}\text{Ti}_{0.96}]_2\text{O}_3$ ceramic	99
18	(a) Coupling factors and (b) dimensional change of $(\text{Pb}_{0.76}\text{Ca}_{0.24})[(\text{Co}_{1/2}\text{W}_{1/2})_{0.04}\text{Ti}_{0.96}]_2\text{O}_3$ ceramics as a function of poling field	101
19	Variation of temperature coefficient of planar resonance frequency for $(\text{Pb,Ln})(\text{Ti,Mn})_2\text{O}_3$ ceramics as a function of Ln concentration	108
20	(a) Coupling factors k_t and k_p and (b) their ratio, k_t/k_p , as a function of ionic radius of Ln for $(\text{Pb}_{0.88}\text{Ln}_{0.08})(\text{Ti}_{0.98}\text{Mn}_{0.02})_2\text{O}_3$ ceramics	109
21	Coupling factors k_p and k_t for $(\text{Pb}_{0.88}\text{Ln}_{0.08})(\text{Ti}_{0.98}\text{Mn}_{0.02})_2\text{O}_3$ ceramic as a function of poling field (Ln=La, Nd, Sm, or Gd.	111
22	The k_p and k_t coupling factors as a function of (a) temperature in $(\text{Pb}_{0.85}\text{Sm}_{0.10})(\text{Ti}_{0.98}\text{Mn}_{0.02})_2\text{O}_3$ ceramics and (b) Sm concentration in $(\text{Pb}_{1-3x/2}\text{Sm}_x)(\text{Ti}_{0.98}\text{Mn}_{0.02})_2\text{O}_3$ ceramics	113
23	X-ray diffraction pattern of calcined and sintered PST powder	121
24	X-ray diffraction pattern of calcined and sintered PCT powder	123
25	Schematic of the sample holder used for piezoelectric resonant measurements	125
26	Conductance and resistance of a typical PST or PCT disk near the planar mode resonance	132
27	Typical positions of the frequencies chosen for calculations of the complex materials coefficients using the Smits' method	137
28	(a) The relative dielectric permittivity and (b) dielectric loss as a function of temperature for PST ceramics near the transition temperature	142
29	(a) The relative dielectric permittivity and (b) dielectric loss as a function of temperature for PCT ceramics near the transition temperature	143

LIST OF FIGURES (cont.)

Figure		Page
30	(a) The relative dielectric permittivity and (b) dielectric loss for PST and PCT ceramics at lower temperatures	144
31	Admittance of a PST disk near the planar mode resonance as a function of poling field at room temperature	147
32	The elastic compliance s_{11}^E for a PST disk at room temperature as a function of poling field	148
33	(a) Poisson's ratio and (b) elastic compliance s_{12}^E for PST ceramics as a function of poling field at room temperature	149
34	(a) The dielectric constant and (b) loss for PST ceramics as a function of poling field at room temperature	150
35	The piezoelectric d_{31} coefficient for PST disks as a function of poling field measured at room temperature	151
36	The electromechanical coupling factors, (a) k_{31} and (b) k_p , for PST disks as a function of poling field, measured at room temperature	152
37	Admittance of a PST disk near the planar mode resonance as a function of temperature	155
38	The series resonant frequencies of the fundamental, $f_{s,1}$, and the first overtone, $f_{s,2}$, harmonic of the planar mode of a PST disk as a function of (a) poling field and (b) temperature	157
39	The elastic compliance s_{11}^E of a PST disk as a function of temperature	159
40	(a) Poisson's ratio and (b) elastic compliance s_{12}^E for PST ceramics as a function of temperature	160
41	(a) The series resonant frequencies of the fundamental, $f_{s,1}$, and the first overtone, $f_{s,2}$, harmonic of the planar mode and (b) the elastic compliance s_{11}^E of a $(\text{Pb}_{0.74}\text{Ca}_{0.26})[(\text{Co}_{1/2}\text{W}_{1/2})_{0.04}\text{Ti}_{0.96}]\text{O}_3$ ceramic disk as a function of temperature	161
42	(a) Poisson's ratio and (b) the elastic compliance s_{12}^E for a $(\text{Pb}_{0.74}\text{Ca}_{0.26})[(\text{Co}_{1/2}\text{W}_{1/2})_{0.04}\text{Ti}_{0.96}]\text{O}_3$ ceramic disk as a function of temperature	162

LIST OF FIGURES (cont.)

Figure		Page
43	Planar coupling coefficient for (a) PST and (b) PCT ceramics as a function of temperature	164
44	Possible temperature dependence of the d_{31} piezoelectric coefficient in PST and PCT ceramics obtained using a standard resonance technique	166
45	(a) Admittance and (b) conductance, of a PST ceramic disk near the fundamental resonant frequency of the thickness mode resonance	170
46	(a) Thickness and planar coupling coefficients and (b) piezoelectric coefficients d_{33} and d_{31} , as a function of poling field in PST ceramics	172
47	The ratio of the (a) thickness and planar coupling coefficients and (b) piezoelectric coefficients, d_{33}/d_{31} , as a function of poling field in PST ceramics	173
48	The series resonant frequencies of the fundamental, $f_{s,1}$, and the first overtone, $f_{s,2}$, harmonic of the thickness mode of a PST disk as a function of temperature	175
49	Planar and thickness coupling coefficients for (a) PST and (b) PCT ceramics as a function of temperature	176
50	Susceptance, B, versus conductance, G, loop for a PST bar at 60°C	180
51	(a) Conductance, G, and (b) susceptance, B, as a function of frequency for a PST bar at 60°C	181
52	(a) The product $d_{31}'d_{31}''$ and (b) the ratio d_{31}''/d_{31}' obtained by the iterative method as a function of temperature for a PST ceramic bar	183
53	Plots of conductance G vs. frequency near the temperature where k_p goes through a minimum for PST ceramics poled at 90 kV/cm	185
54	Possible temperature dependence of the real and imaginary components of the complex d_{31} piezoelectric coefficient in PST and PCT ceramics obtained using the iterative method	188

LIST OF FIGURES (cont.)

Figure		Page
55	(a) The real and (b) imaginary components of complex d_{31} as a function of temperature for a PST bar poled at 30 kV/cm	190
56	(a) The real and (b) imaginary components of complex s_{11}^E as a function of temperature for a PST bar poled at 30 kV/cm	191
57	(a) The real and (b) imaginary components of complex ϵ_{33}^x as a function of temperature for a PST bar poled at 30 kV/cm	192
58	Real part of the piezoelectric coefficient, d_{31} , at different poling fields as a function of temperature for PST ceramics	194
59	(a) Real part of the piezoelectric coefficient, d_{31} , and (b) electromechanical coupling factor, k_{31} , as a function of poling field for PST ceramics at 20°C	196
60	Real part of the piezoelectric coefficient, d_{31} , and the transverse electromechanical coupling factor, k_{31} , as a function of poling field for PST ceramics at various temperatures	197
61	(a) Complex piezoelectric coefficient d_{31} and (b) electromechanical coupling factor k_{31} , as a function of temperature for PCT ceramics poled at 50 kV/cm	208
62	Real part of the piezoelectric coefficient d_{31} of PST ceramics measured as a function of frequency at different poling fields using the interferometric technique	210
63	Planar coupling factor for PST ceramics and a highly conductive Sm-modified lead titanate ceramic as a function of temperature	213
64	Planar coupling factor for Sm-modified lead titanate ceramics, prepared using a sol-gel process, as a function of temperature at different sintering times	214
65	The transverse mechanical quality factor, $Q=s'/s''$, for PCT ceramics as a function of temperature	220
66	(a) The ϵ'/ϵ'' and (b) d'/d'' ratios for PCT ceramics as a function of temperature	221
67	Real part of the piezoelectric coefficient d_{31} for a $(\text{Pb}_{0.66}\text{Ca}_{0.34})\text{TiO}_3$ ceramic as a function of temperature	223