

Electrostriction in Perovskite Crystals and Its Applications to Transducers

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Electrostrictive materials for displacement transducers are reviewed, including preparative, theoretical, and design studies. Several empirical rules have been deduced from investigation of electrostrictive effects in ferroelectric, antiferroelectric, and non-polar perovskites. Consistent with these criteria, the relaxor ferroelectric 0.9Pb(Mg_{1/3}Nb_{2/3})O₃-0.1PbTiO₃ has been shown to possess much larger strain with lower hysteresis, aging effects, and thermal expansion than commercial piezoelectric PZT. Using a multilayer configuration, we have developed a mirror-control device capable of large strains up to $\Delta L/L \sim 10^{-3}$ with only 200V applied.

Recent requirements for new displacive transducers with lower hysteresis and aging effects than conventional PZT based ceramics have prompted the development of a new family of electrostrictive ceramics for these applications.¹⁾ In this paper we review the properties of electrostrictive transducers, including recent experimental and theoretical studies.

Electrostriction is a measure of the strain resulting from ionic displacements caused by an applied electric field. In most centric crystals, the induced shifts of equivalent ions almost cancel one another, with only the differences arising from anharmonicity creating strain. It is thus not unreasonable to expect interrelations between anharmonic effects such as thermal expansion and electrostriction. Figure 1 shows the thermal expansion coefficient α plotted against the hydrostatic electrostriction

coefficient $Q_h (= Q_{11} + 2Q_{12})$ for materials of isotropic or cubic symmetry. The power relation: $\alpha = 4.2 \times 10^{-5} Q_h^{0.5}$ was obtained from this graph (empirical rule I).²⁾

There are several experimental methods for determining the electrostriction coefficients. Direct measurements of field-induced or spontaneous strain include optical methods (interferometer,³⁾ optical lever, optical grids), x-ray methods, electrical methods (capacitance dilatometer,^{4,5)} differential transformer dilatometer^{6,7)} and strain gauge methods.^{8,9)} Pressure gauge methods, measurements of the permittivity variation with pressure^{8,10)} and induced piezoelectric resonance techniques^{3,11)} are indirect methods.

In Table I, hydrostatic electrostriction coefficients Q_h and Curie-Weiss constants C are summarized for various kinds of oxide perovskite crystals: simple, disordered, partially ordered and ordered ferroelectrics, antiferroelectrics and non-polar dielectrics. Sources of the original experimental data are listed in references 8, 10, and 12. The magnitude of the electrostriction coefficient is not affected strongly by ferroelectricity, antiferroelectricity, or non-polar behavior, but is very dependent on the degree of order in the cation arrangement. An empirical rule II is proposed that states "the electrostrictive Q coefficient increases with cation order from disordered, through partially ordered, simple and then ordered perovskites." The decrease in the Curie-Weiss constant with increase in the degree of cation ordering is also

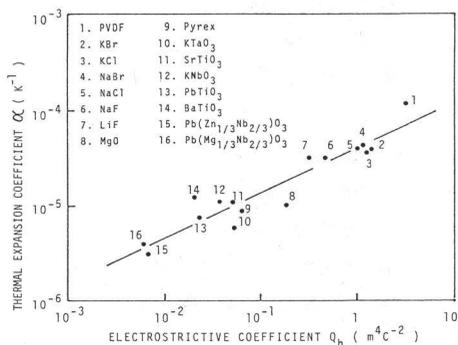


Fig. 1. Thermal expansion coefficient α plotted as a function of hydrostatic electrostriction coefficient Q_h for various materials. The straight line has slope 0.5.

Table I. Electrostrictive coefficients, Curie-Weiss constants and their product values for various perovskite-type crystals.

Polar-Type	Order-type	Substance	Q_h ($\times 10^{-2} \text{m}^4 \text{C}^{-2}$)	C ($\times 10^5 \text{K}$)	$Q_h C$ ($\times 10^3 \text{m}^4 \text{C}^{-2} \text{K}$)	References
Ferroelectric	Disordered	$\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$	0.60	4.7	2.8	Uchino (1980a), Nomura (1979a)
		$\text{Pb}(\text{Zn}_{1/3}\text{Nb}_{2/3})\text{O}_3$	0.66	4.7	3.1	Nomura (1979b), Kuwata (1979)
	Partially ordered	$\text{Pb}(\text{Sc}_{1/2}\text{Ta}_{1/2})\text{O}_3$	0.83	3.5	2.9	Setter (1980)
	Simple	BaTiO_3	2.0	1.5	3.0	Yamada (1972), Johnson (1965)
		PbTiO_3	2.2	1.7	3.7	Gavrilyachenko (1970), Fesenko (1970)
		SrTiO_3	4.7	0.77	3.6	Samara (1966), Bell (1963)
KTaO_3		5.2	0.5	2.6	Uwe (1975), Uwe (1973)	
Antiferroelectric	Partially ordered	$\text{Pb}(\text{Fe}_{2/3}\text{U}_{1/3})\text{O}_3$	—	2.3	—	Uchino (1977)
	Simple	PbZrO_3	2.0	1.6	3.2	Uchino (1980b), Samara (1970)
	Ordered	$\text{Pb}(\text{Co}_{1/2}\text{W}_{1/2})\text{O}_3$	—	1.2	—	Bokov (1965)
$\text{Pb}(\text{Mg}_{1/2}\text{W}_{1/2})\text{O}_3$		6.2	0.42	2.6	Uchino (1980b), Nomura (1979a)	
Non-polar	Disordered	$(\text{K}_{3/4}\text{Bi}_{1/4})(\text{Zn}_{1/6}\text{Nb}_{5/6})\text{O}_3$	0.55–1.15	—	—	Uchino (1980c)
	Simple	BaZrO_3	2.3	—	—	Uchino (1980c)

correlated with electrostriction. This leads to an empirical rule III, “the product of the electrostriction coefficient Q and the Curie-Weiss constant C is nearly constant for all ferroelectric and antiferroelectric perovskites ($Q_h C = 3.1(\pm 0.4) \times 10^3 \text{m}^4 \text{C}^{-2} \text{K}$).” We have proposed an intuitive crystallographic model to explain the “constant QC rule.” Figures 2(a) and (b) show the ordered and disordered structures for an $A(B_{I1/2}B_{II1/2})\text{O}_3$ perovskite crystal.

Assuming a rigid ion model, a large “rattling” space is expected for the smaller B ions in the disordered structure because the larger B ions prop open the lattice framework. Much less rattling space is expected in the ordered arrangement where neighboring ions collapse systematically around the small B ions.¹³⁾ When an electric field is applied to a disordered perovskite, the B ion with large rattling space can shift easily without distorting the oxygen framework. Larger polarization can be expected for unit magnitude of electric field, causing

larger dielectric constants and larger Curie-Weiss constants. Under these circumstances,

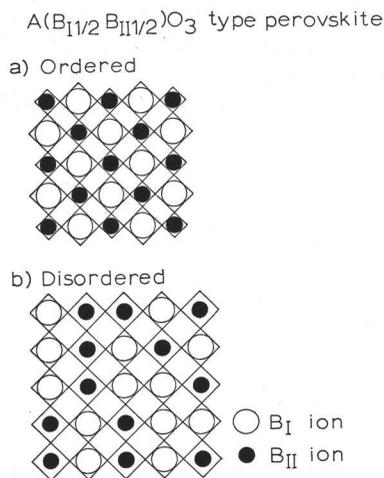


Fig. 2. Crystal structure models of the $A(B_{I1/2}B_{II1/2})\text{O}_3$ type perovskite:

- (a) Ordered structure with a small rattling space.
(b) Disordered structure with a large rattling space.

smaller strains are expected per unit magnitude of polarization, resulting in lower electrostriction coefficients. On the other hand, in ordered perovskites with very small rattling space, the B ions cannot move easily without distorting the oxygen octahedron. A smaller Curie-Weiss constant and a larger electrostriction coefficient are expected. The measurements on solid solution systems are also very suggestive. The results for $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-PbTiO}_3$,^{6,7)} $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-Pb}(\text{Mg}_{1/2}\text{W}_{1/2})\text{O}_3$ ⁹⁾ and $\text{SrTiO}_3\text{-Bi}_{2/3}\text{TiO}_3$ ¹⁰⁾ indicate that the electrostriction coefficients decrease substantially with increasing rattling space.

Since the figure of merit for electrostrictive strain is $Q\varepsilon^2$ (ε : permittivity) or QC^2 (eliminating temperature dependence), and the product QC is nearly constant for all ferroelectric perovskites, disordered perovskites with large Curie-Weiss constants, but small electrostriction coefficients, are preferred to the usual perovskites (e.g. $\text{Pb}(\text{Zr}, \text{Ti})\text{O}_3$ or BaTiO_3 based ceramics) for practical applications. The relaxor ferroelectric chosen for study is $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$, which itself is superior to conventional modified BaTiO_3 ceramics in its electrostrictive response.⁷⁾ The response can be further improved if the Curie range, which is below room temperature in $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$, is shifted to slightly higher temperature. The Curie range of the solid solution $0.9\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-}0.1\text{PbTiO}_3$ extends from 0°C to 40°C . Using a differential transformer dilatometer, the transverse electrostrictive strain was measured along the length of a thin ceramic rod, subject to DC bias fields applied in a perpendicular direction (Fig. 3).⁶⁾ The relaxor ceramics are anhysteretic and retrace the same curve with rising and falling fields. For comparison the piezoelectric strain of a hard PZT 8 under cyclic fields is also plotted in Fig. 5. This material has often been used in the fabrication of active optical components.¹⁴⁾ Field-induced strains in $0.9\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-}0.1\text{PbTiO}_3$ are larger than those in PZT and far more reproducible under cyclic drive conditions. An additional merit is that electric poling is not required.

Another interesting property of relaxor ferroelectrics is the very small thermal expansion effect throughout the Curie range,

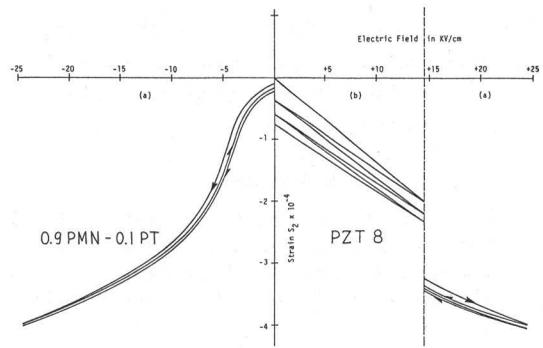


Fig. 3. Transverse strain in ceramic specimens of (a) $0.9\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-}0.1\text{PbTiO}_3$, and (b) a typical hard PZT 8 piezoceramic under slowly varying electric fields.

as predicted from empirical rule I ($\alpha^2 \propto Q$). The thermal expansion coefficient of $0.9\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-}0.1\text{PbTiO}_3$ in the temperature range -100°C to $+100^\circ\text{C}$ is less than $1 \times 10^{-6}\text{K}^{-1}$, comparable to the best low expansion ceramics or fused silica. The thermal strains are far smaller than the electrostrictive strains, which is extremely advantageous for micropositioner applications.

The multilayer technology used in the capacitor industry is one of the important factors prompting the development of new electrostrictive devices. The electric field across alternate layers is of opposite direction, but the displacive responses are additive. In a piezoelectric device of fixed total thickness, the total displacement for a given voltage is proportional to the number of layers. On the other hand, in an electrostrictive device the total displacement is proportional to the square of the number of layers, far more effective than in a piezoelectric material. Internally electroded ten-layer devices with a total thickness of 2.5 mm were prepared by standard tape casting techniques using calcined $0.9\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-}0.1\text{PbTiO}_3$ powder and a commercial doctor blade medium.^{15,16)} A mirror control device has been constructed from 10 ten-layer ceramics bonded together. With only 200V applied, the device develops large displacements up to $\Delta L \sim 25\ \mu\text{m}$ ($\Delta L/L \sim 10^{-3}$) with very high reproducibility under cyclic fields. The total displacement of about $25\ \mu\text{m}$ is an order of magnitude larger than that of commercial piezoelectric transducers (e.g. PZT-5H, 25 mm plate using d_{31}),¹⁷⁾ and may introduce a new class of

micropositioner devices. With further refinement of the tape casting process, it is probable that the driving voltage can be further reduced to less than 40V.

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