INTRODUCTION

Relaxor electrostrictors such as those based on Pb(Mg\textsubscript{1/3}Nb\textsubscript{2/3})\textsubscript{O}\textsubscript{3}-PbTi\textsubscript{O}\textsubscript{3} (PMN-PT) have desirable properties which are unavailable in conventional piezoelectric materials. Foremost is a high induced strain with low hysteresis at reasonable drive fields, over 0.1\% strain at 1.0 MV/m with less than 5\% electromechanical hysteresis in some compositions\textsuperscript{1}. Unlike the linear response to drive fields observed in piezoelectrics, electrostrictors show a quadratic response with strain values high enough to make them useful as engineering materials.

Typical relaxor electrostrictors demonstrate a diffuse transition region between high temperature electrostrictive properties and low temperature, primarily piezoelectric, properties. Weak-field dielectric properties indicate a temperature at which the dielectric constant is a maximum for a given frequency, but because of the diffuse nature and possible field dependence of the transition this is not necessarily the transition temperature for relaxors in high fields. The effective electromechanical Q is a candidate parameter for determining a reliable transition temperature for high-field properties. The quantity $Q_{\text{eff}}$, the effective electromechanical Q, is defined as the peak induced strain for a given electric field divided by the square of the induced polarization. Preliminary results show that $Q_{\text{eff}}$ is useful as a parameter for indicating the transition temperature of PMN-PT-X electrostrictors in high fields, confirming a previous study\textsuperscript{1}.

Some other issues under investigation are the influence of barium and strontium titanates, as dopants for PMN-PT, on the phase transition properties, the characteristics of phase transition properties as a function of field, and the possibility of extrapolating low frequency behavior to predict high frequency response.

The compositions chosen for this investigation show high strains with low hysteresis near room temperature. The compositions are 0.975(0.925Pb(Mg\textsubscript{1/3}Nb\textsubscript{2/3})\textsubscript{O}\textsubscript{3}-0.075PbTi\textsubscript{O}\textsubscript{3})-0.025BaTi\textsubscript{O}\textsubscript{3}, and 0.975(0.910Pb(Mg\textsubscript{1/3}Nb\textsubscript{2/3})\textsubscript{O}\textsubscript{3}-0.090PbTi\textsubscript{O}\textsubscript{3})-0.025BaTi\textsubscript{O}\textsubscript{3}. These are members of the PMN-PT-X family, where PT signifies PbTiO\textsubscript{3} and X is BaTiO\textsubscript{3}. The compositions are designated b250075 and b250090, respectively. Similar compositions with SrTiO\textsubscript{3} as a dopant are being studied in continuing work.

The fabrication of the compositions has been described elsewhere using columbite as a precursor powder\textsuperscript{2}. The method was modified by using isopropyl alcohol as the milling agent. Mixing and grinding were accomplished by vibratory milling, employing commercially available raw materials. The finished pellets formed were approximately 8 mm in diameter and 1.5 mm thick. Perovskite phase purity was determined by X-ray diffractometry. All samples showed no pyrochlore to the limits of detection. Electrodes were formed on the pellets with sputtered gold and fired-on silver.

Weak-field (1 Volt AC) properties were collected for relative permittivity and dielectric loss between -40 and 80\textdegree C at frequencies of 0.1, 1, 10, and 100 kHz with an impedance analyzer and a computer controlled environmental chamber. For both increasing and decreasing temperature, frequency dispersion typical of relaxor ferroelectric materials was observed with no dielectric aging.

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Figure 1 shows the weak-field properties of b250075. For each frequency, the maximum permittivity and the temperature at which it occurred, $T_{\text{max}}$, were determined, as well as the associated loss tangent.

The transverse strain and polarization of the compositions were measured at 10, 100 and 200 Hz. Induced transverse strain and polarization were measured simultaneously. These high-field properties were acquired between -40 and 80°C with drive fields of 0-0.5 MV/m (0.25 MV/m DC bias) and 0-1.0 MV/m (0.5 MV/m DC bias) at 10 Hz. Measurements were taken using a circuit modelled after Sawyer and Tower,3 with a large integrating capacitor, to determine polarization. Strain gauges bonded to the sample electrodes were used to measure the induced transverse strain. A multichannel digitizing oscilloscope and a computer collected the data.

**DISCUSSION**

Relaxors in the paraelectric (exclusively electrostrictive) state are generally regarded as possessing a quadratic variation of strain with polarization, with a coefficient, $Q$, independent of temperature and field.4 Since the measurements in this study include temperatures below those required for paraelectric behavior for the given composition, a piezoelectric contribution must be considered for any correlations between strain and polarization. A measurement of an effective transverse electromechanical $Q$, $Q_{\text{eff}}$, can be made by dividing the peak induced transverse strain by the square of the peak induced polarization for a given drive field, as described by the following simplified formula:

$$Q_{\text{eff}} = \frac{s}{P^2} = \frac{xg}{P} + xQ'_{12} + (1-x)Q_{12},$$

where $s$ is the peak induced transverse strain, $P$ is the peak polarization, $x$ is the volume of material demonstrating piezoelectric behavior, $g$ is the piezoelectric coupling factor, and $Q'_{12}$ and $Q_{12}$ are the electrostrictive $Q$ coefficients for the respective piezoelectric and paraelectric phases. $Q_{\text{eff}}$ is therefore field and frequency dependent below the temperature at which the relaxor electrostrictor begins to show piezoelectric character in any part of its volume. $Q_{\text{eff}}$ is independent of field and frequency above the temperature where piezoelectric behavior is lost.

Figure 2 is a plot of $Q_{\text{eff}}$ and temperature at 10 Hz for both 0-0.5 MV/m and 0-1.0 MV/m. Above 25°C, $Q_{\text{eff}}$ is constant within experimental error, which indicates that the relaxor is in its paraelectric phase. No frequency or field dependence can be observed, as expected. Between 15 and 25°C a change in the value of $Q_{\text{eff}}$ with temperature is discernible for both field strengths. At lower temperatures a field dependence is apparent, indicating the influence of a piezoelectric region in the material.

The linear relationship between $T_{\text{max}}$ and frequency, which can be observed in Figure 1, indicates that $T_{\text{max}}$ for b250075 at 10 Hz is about 5°C. Similarly, $T_{\text{max}}$ at 10 Hz for b250090 is about 15°C. $T_{\text{max}}$ can also be found for other frequencies. In Figure 1, the dielectric loss at $T_{\text{max}}$ for any given frequency can be seen to be significantly above the minimum value at higher temperatures. This indicates that a volume of the material is in the piezoelectric state at $T_{\text{max}}$.

The weak-field properties discussed above indicate a $T_{\text{max}}$ of about 5°C for b250075. The dielectric loss at $T_{\text{max}}$ for any given frequency is significantly above the minimum value found at higher temperatures, suggesting that a volume of the material is in its piezoelectric state at $T_{\text{max}}$. Since the transition temperature calculated from $Q_{\text{eff}}$ for b250075 at 10 Hz is 15°C, i.e. 10°C higher than $T_{\text{max}}$, the two values are in accordance with the existence of some piezoelectric component at $T_{\text{max}}$. Similarly, $Q_{\text{eff}}$ changes from a constant above approximately 30°C at 10 Hz in b250090, and $T_{\text{max}}$ is approximately 20°C. This behavior also occurs at 100 and 200 Hz for both compositions.
CONCLUSIONS

Preliminary data presented here for the relaxor electrostrictors b250075 and b250090 show that $Q_{\text{eff}}$ indicates a transition temperature in PMN-PT-X materials between the purely electrostrictive state and the piezoelectric state, occurring in small volumes of the material, at various field strengths. Further work is continuing in the examination of the induced strain and polarization, and hysteresis behavior of b250075 and b250090 as a function of frequency, field, and temperature. Other relaxor ferroelectrics in the PMN-PT-X family, where $X$ will be SrTiO$_3$, will be included in the study.

REFERENCES


Figure 1. Weak field dielectric properties of the relaxor ferroelectric composition $0.975(0.925\text{Pb(Mg}_{1/3}\text{Nb}_{2/3})\text{O}_{3}-0.075\text{PbTiO}_{3})-0.025\text{BaTiO}_{3}$. The dashed line indicates the linear relation between $T_{\text{max}}$ and frequency.
Figure 2. $Q_{eff}$ at 10 Hz for b250075 at 0-0.5 MV/m and 0-1.0 MV/m. Note the field invariance at high temperatures and the divergence at low temperatures.