Relaxor-PT Single Crystals: Observations and Developments

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Abstract — Relaxor-PT single crystals are at the forefront of advanced transducer applications, including medical ultrasound and high power sonar. In this work, we report on recent developments in Relaxor-PT crystals, specifically increased temperature usage and crystallographic and domain engineering relationships. Furthermore, we contrast developments in crystals with respect to both "soft" and "hard" piezoelectric ceramics, namely $Pb(Zr,Ti)O_3$ (PZT).

INTRODUCTION

Recently, extensive research has been carried out on relaxor-PT ferroelectric single crystals. As reported, domain engineered $Pb(Zn_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ (PZNT) and $Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ (PMNT) crystals oriented along the pseudo-cubic <001> direction exhibit high electromechanical coupling factors and piezoelectric coefficients for the longitudinal mode, with k_{33} factors on the order of > 0.90 and d_{33} higher than 1500pC/N [1]. However, their usage temperature range is limited by their relatively low Curie temperature (T_C ~130-170°C) and further restricted by their lower ferroelectric phase transition temperature, being on the order of T_{R-T} ~ 60-100°C, the consequence of a strong curvature in their morphotropic phase boundary (MPB). Furthermore, relaxor-PT crystals exhibit low mechanical quality factors Qs (<100) and coercive fields (E_C ~2-3 kV/cm), showing typical "soft" characteristics, limiting their application in high power transducers. Thus, numerous investigations have been carried out to explore new relaxor-PT single crystal systems, where higher Curie temperatures and high coercive fields and high mechanical Qs are desired [2-4].

In this work, we summarized general observations and recent developments of relaxor-PT single crystal systems, with general relationships highlighted in the following: 1) Electromechanical coupling factors vs transition temperatures; 2) piezoelectric coefficients and dielectric permittivity vs T_{R-T}; 3) Coercive field vs Curie temperature; 4) Mechanical quality factor Q vs electromechanical coupling.

EXPERIMENTAL

Relaxor-PT single crystals, including PMNT, PMN-PIN-PT and PMN-PZ-PT were grown using the conventional Bridgman growth method and solid state crystal growth (SSCG), respectively. Other ferroelectric crystals, such as $BiScO_3$-$PbTiO_3$, were obtained by high temperature solid solution method. The crystals were oriented along various crystallographic directions <001>, <110> and <111> and made into different samples with geometries following the IEEE standard on piezoelectricity. The samples were vacuum sputtered with gold thin films as electrodes and poled at electric field of 10-20kV/cm, depending on their related coercive field. The capacitance, resonance and anti-resonance frequencies for different vibration modes were measured using multi-frequency LCR meter (HP4284A) and HP4194A Impedance- phase gain analyzer, respectively, from which, the dielectric permittivity, elastic constant, electromechanical coupling factor, piezoelectric coefficient and mechanical quality factor could be calculated. The Curie temperature and T_{RT} were obtained from the dielectric temperature behavior at 1kHz frequency. The coercive field E_C and internal bias E_i were recorded from the polarization hysteresis (P-E loop) measured by the modified Sawyer-Tower circuit, at 1Hz frequency and electric field of 10-40kV/cm, where the PE loops should be in saturated shape, prior to its electric field induced phase transition.

RESULTS AND DISCUSSIONS

Fig. 1 shows the general relationship of electromechanical coupling factor k_{33} for relaxor-PT single crystals as a function of temperature, where it was found that coupling is independent of the Curie temperature and/or ferroelectric phase transition temperature, being on the order of 0.9 for most perovskite ferroelectric crystals with MPB compositions. Fig. 2 &3 give the relationship of dielectric permittivity and piezoelectric coefficient d_{33} as a function of ferroelectric phase transition temperature T_{RT}. As shown, the values decrease with increasing T_{RT}, similar to the T_C dependence of PZTs.

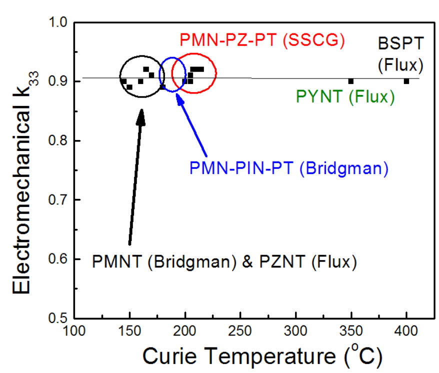

Fig. 1 Electromechanical coupling k_{33} as function of T_C.

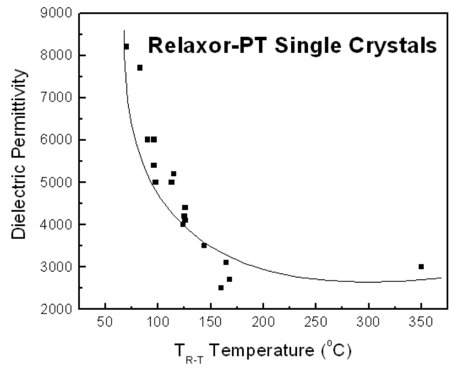

Fig. 2 Dielectric permittivity as function of T_{RT}.

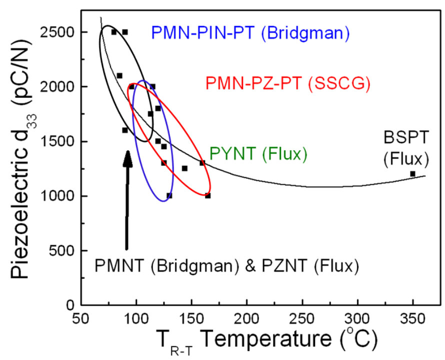

Fig. 3 Piezoelectric coefficients as function of T_{RT}.

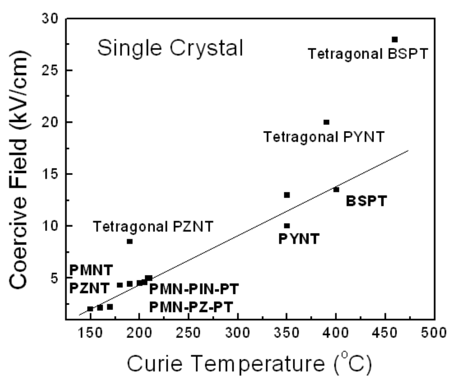

Fig. 4 Coercive field as function of Curie temperature.

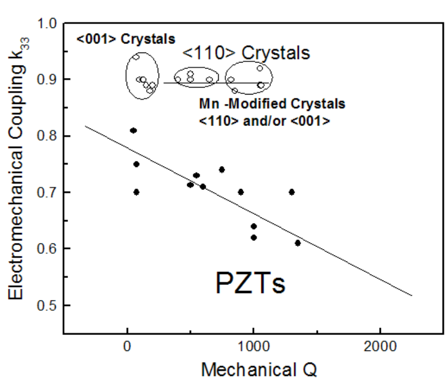

Fig. 5 The relationship between Electromechanical coupling and mechanical Q for crystals and ceramics.

Fig. 4 exhibits the trend of coercive field as a function of Curie temperature for relaxor-PT single crystals, where it was found the coercive field increased with increasing T_C. The coercive field was found to be on the order of 2-3kV/cm for PMNT crystals, increasing to 5kV/cm for modified PMNT, with Curie temperatures 30-80^{o}C higher than the pure PMNT, further increased to 13kV/cm for BSPT crystal with a Curie temperature on the order of >400^{o}C. Also, tetragonal phase crystals were found to possess much higher coercive fields when compared to their rhombohedral phase counterparts. Fig. 5 shows the coupling k_{33} as function of mechanical Q, where it was found that high k_{33} are for "soft" (low Q) PZT ceramics, decreasing for "hard" (high Q) PZTs. However, as can be found in Fig. 1, the k_{33} maintains similar values of 0.9 for perovskite crystals, while the Q values can be tuned using the acceptor dopants and/or domain engineering, being in the range of 70-1000 [3,4].

SUMMARY

In summary, the properties of relaxor-PT single crystals, including dielectric permittivity, piezoelectric coefficients, electromechanical coupling, coercive field and mechanical quality factor were investigated as a function of Curie temperature T_c and/or T_{RT}, where it was found the crystals behave differently from polycrystalline ceramics. The dielectric and piezoelectric properties reduced with increasing T_{RT}, and not T_c, while the coercive field increased with increasing T_C for both crystals and ceramics. Of particular interest is that the electromechanical coupling k_{33} was found to be ~0.9, regardless of the T_C. Mechanical Qs could range from 70 to 1000, depending on acceptor dopant and domain engineering configuration.

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