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Thickness Dependent Characteristics of High Permittivity PMN-0.32PT Single Crystal for High Frequency Medical Imaging Applications

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Abstract

The dielectric, piezoelectric, and acoustic properties of PMN-0.32PT ($\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-PbTiO}_3$) single crystals were investigated as a function of sample thickness ranging from 120 to 30 μm in order to enlighten the origin of property degradation of crystals for high frequency ultrasound applications. Electromechanical coupling factor(k_t), clamped and free dielectric constants decreased but sound velocity increased with decreasing crystal thickness. Particularly, repoling of the PMN-PT crystals would bring about a noteworthy enhancement in electromechanical and dielectric properties, which urges the importance of PMN-PT as a promising piezoelectric material for high frequency ultrasound transducers.

Keywords

PMN-PT; piezoelectric property; dielectric property; thickness dependence

I. Introduction

Ultrasonic imaging has been widely used in medical imaging applications due to their non-invasive nature and superior capabilities in acquiring information of living tissues [1]. A piezoelectric material is an important component in an ultrasound transducer [2]. Among the wide range of piezoelectric materials, recent advances in developing single crystal relaxor ferroelectrics, such as $(1-x)\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-}x\text{PbTiO}_3$ (PMN-PT) have attracted much attention due to their high dielectric permittivity ($\epsilon_r > 5500$) and high electromechanical coupling coefficient ($k_{33} \sim 0.92$) [3, 4, 5]. In addition, single crystals do not have the grain-size issues as for polycrystalline ceramics at higher frequencies (>20 MHz). PMN-PT crystal is a good alternative for high frequency ultrasound transducers/arrays [6, 7].

So far, various aspects of the thickness dependence of the PMN-0.32PT have not been investigated. Recent works for PMN-PT single crystal have reported a decrease in electromechanical coupling coefficient as the thickness is reduced. This observation

prompted an investigation on dependency of other characteristics with thickness in PMN-PT materials [8].

This research is intended to characterize the dielectric, piezoelectric and acoustic properties of PMN-PT single crystal as a function of sample thickness/corresponding frequency in order to enlighten the origin of property degradation of crystals for high frequency ultrasound applications. In addition, a method to compensate the deterioration of some characteristics in higher frequencies has been presented and tested.

II. Experimental

PMN-PT single crystals were obtained from HC Materials Corporation (Bolingbrook, IL). PMN-PT samples with different thicknesses were lapped and polished on both sides. Initial lapping utilized 12 micron alumina abrasive powder and distilled water. Final polishing was performed using 3.0 micron and 0.3 micron alumina paste. Cr/Au (500 Å/1000 Å) electrodes were deposited onto both sides. Samples were then cut to specific dimensions with an aspect ratio (width/thickness) greater than 20. The large aspect ratio helps to reduce interference of undesired lateral mode with the fundamental thickness mode [9]. The prepared samples were all annealed at 400 °C for 4 hours for stress releasing prior to the sputtering step. The samples were poled at electric field of 3 V/μm for 5 min at room temperature [10].

The dielectric properties of the samples were measured in air using an Agilent 4294A precision impedance analyzer. The free and clamped permittivity (K_{33}^T, K_{33}^S) were calculated from the low (1 kHz) and high frequency (about twice as anti-resonance frequency, f_a) capacitances respectively according to the equation [11]:

$$K_{33} = \frac{Cd}{A\epsilon_0}$$

where C is the capacitance, d is the thickness of the element, A is the area of the element, ϵ_0 is 8.85×10^{-12} f/m. The thickness mode electromechanical coupling factor (k_t), were calculated from the resonance and anti-resonance frequencies of resonators according to the IEEE standards using following equation [12]:

$$k_t = \sqrt{\frac{\pi}{2} \frac{f_r}{f_a} \tan\left(\frac{\pi}{2} \frac{f_a - f_r}{f_a}\right)}$$

The longitudinal sound velocity (v_l) was calculated from the measured resonance frequency (f_r) and sample thickness. Finally, the effect of electrical field during poling on the electromechanical and dielectric properties at higher frequencies was studied.

III. Results and Discussion

Figure 1 illustrates the thickness dependence of thickness mode electromechanical coupling factor (k_t) with two 3.0 μm and 0.3 μm finishing. Crystal thicknesses range from 120 μm to 30 μm. With increased corresponding resonance frequency the electromechanical coupling factor (k_t) decreased from 0.59 to 0.48 for 0.3 μm and from 0.56 to 0.44 for 3.0 μm finishing respectively. Samples with finer surface finishing (0.3 μm) have k_t values about 8% larger than those of the coarser surface samples (3.0 μm). In both cases, coupling coefficient (k_t) exhibited a decreasing trend with crystal thickness (increasing resonance frequency).

Thickness dependant plots of free (K_{33}^T) and clamped (K_{33}^S) dielectric constants are given in Fig. 2, and Fig. 3. PMN-PT samples exhibit a reduction in both free and clamped dielectric constants from 7102 to 4776, and 832 to 695 respectively as crystal part thickness decreases from 120 μm to 30 μm .

The free dielectric constant presents a steeper reduction than the clamped one and it appears to be more sensitive to the crystal thickness.

The property degradation in ferroelectric materials when sample thickness approaches the size of domains would be attributed to the surface boundary defects which may bring about non-ferroelectric or low polarizable layers [13]. Such degradation could also be due to corresponding large domain size, domain clamping and polarization rotation dampening [9, 14].

The longitudinal velocity, on the contrary, increased from 4108 to 4771 m/s with decreasing sample thickness (Fig. 4). The increase in velocity with decreasing thicknesses could be associated with the hardening of layers; which happens due to the residual stress produced by the cold work resulted by lapping. This behavior of sound velocity can also be brought about by plane dislocations, and vacancy concentration between atoms. A higher vacancy concentration leads to an increase in micro-hardness, and a lower compressibility (G) which inherently results in an increase in sound velocity (c) [11].

Of particular interest is that repoling of PMN-PT samples (30 μm thick with 0.3 μm finish) appears to bring about a significant improvement in the electromechanical coupling factor (k_t) from 0.48 to 0.55 (Fig. 5), as well as the free and clamped relative dielectric constants from 4776 to 5812 (Fig. 6) and 701 to 761 (Fig. 7) respectively. This behavior suggests repoling after lapping in order to recover their electromechanical and dielectric properties for higher frequency applications. However, it couldn't fully recover to previous data ($k_t \sim 0.58$). It may be caused by stress effect [11].

IV. Conclusion

In the present paper, we studied the thickness dependence of electromechanical, dielectric, and acoustic properties for PMN-0.32PT single crystal. Crystals showed a reduction in the electromechanical coupling factor (k_t), as well as free and clamped relative dielectric constants with decreasing crystal thickness. A smoother surface finishing yielded increased k_t as the part has less surface damages. Moreover, sound velocity was found to increase with increasing frequency which might be associated with the increasing domain size, and micro-hardening which could lead to an increase in compressibility and sound velocity. Of the paramount importance is repoling of the samples which enhance both electromechanical coupling and dielectric properties significantly, allowing PMN-PT single crystals to remain attractive for high frequency, and broad band transducer applications.

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Figure 1.
Thickness dependence of electromechanical coupling factor.

Figure 2.
Frequency dependence of free dielectric constant.

Figure 3.
Frequency dependence of clamped dielectric constant.

Figure 4.
Frequency dependence longitudinal sound velocity.

Figure 5.
Repoling effect on the electromechanical coupling factor.

Figure 6.
Repoling effect on free dielectric constant.

Figure 7.
Repoling effect on clamped dielectric constant.