Status of Piezoelectric Single Crystal Growth for Medical Transducer Applications

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Abstract – Single crystal piezoelectrics such as PMN-PT are leading to dramatic improvements in transducer, sensor and actuator technology. The potential benefit of implementing these materials is clearly promising in terms of increased bandwidth, sensitivity and source levels due to ultrahigh electromechanical coupling factors (k_{33} >94%), high piezoelectric coefficients (d_{33} >2500pC/N), high strain levels (>0.6%), and low hysteresis.

TRS is investigating a variety of uses for piezoelectric single crystals in medical transducer applications such as ultrasound harmonic imaging and high frequency transducers. Harmonic imaging requires very broad bandwidth ultrasonic transducers. By using single crystals, which inherently have more bandwidth, the complex engineering normally associated with achieving increased bandwidth will be reduced. Simple ultrasound transducers that have been constructed from crystals exhibiting bandwidths of 100% (if high impedance, backing layers are used, 140% can be achieved) and insertion losses less than similarly constructed PZT transducers with only a 70% bandwidth.

To date, commercialization of these materials for ultrasound has been limited by low yields, small size, and high cost. The primary cost driver for single crystals is the amount of useable material yielded per growth run. Currently these crystals are grown by the Bridgman method, which involves moving a crucible containing a molten charge relative to a stationary temperature gradient resulting in unidirectional solidification. One limiting factor of the Bridgman growth technique for PMN-PT is that useful compositions of this system have a segregation coefficient less than 1, so there is a significant compositional gradient along the growth axis, which severely limits the amount of crystal yielded per growth run. The current state of the art is Bridgman-grown 1.5-2" diameter boules. Scaling up and the use of zone melting in the Bridgman technique to increase useable length are expected to yield price reductions for large volume production that are 2 to 3 times that of high quality PZT ceramic (~$20 to 30/cc).

The current status of the growth development including current issues as well as preliminary results of utilizing crystals in medical transducer applications will be discussed.

I. SINGLE CRYSTAL TRANSDUCERS

Single crystals based on $(1\text{-}x)Pb(Zn_{1/3}Nb_{2/3})O_3\text{-}xPbTiO_3$ (PZN-PT) or $(1\text{-}x)Pb(Mg_{1/3}Nb_{2/3})O_3\text{-}xPbTiO_3$ (PMN-PT) represent a revolutionary advance in piezoelectric transducer technology. These materials exhibit 5 to 10 times the strain of conventional piezoelectric ceramic with high electromechanical coupling. Therefore, they offer a potential for higher performance transducers with broad bandwidth. The most important parameter affecting bandwidth is the electromechanical coupling factor of the piezoelectric resonator. The current material of choice for broadband transducers is Type VI PZT ceramic with a longitudinal coupling (k_{33}) of 0.75 to 0.80. However, since transducers are usually fabricated for beam mode operation (see Figure 1) the actual coupling coefficient tends to be closer to 0.70. This was long thought to be the best possible piezo-

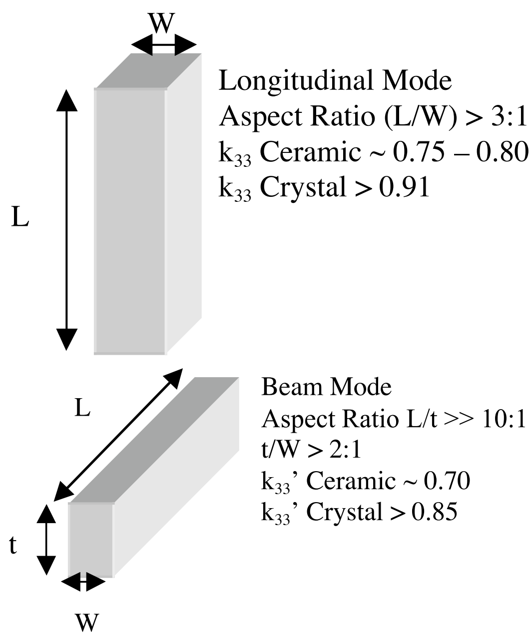

Figure 1: Piezoelectric resonators for ultrasound transducers: a) longitudinal mode resonator characterized by k_{33} coupling coefficient; used in 1-3 composites and 1.5-D phased arrays, b) beam mode resonator characterized by k_{33}' coupling coefficient (< k_{33}); used in 2-2 composites, linear arrays, and 1-D phased arrays.

Table I: Properties of Single Crystal Piezoelectrics Compared to Ceramics Commonly used for Imaging and Therapeutic Ultrasound.

Property	Type VI PZT (TRS610)	PMN-33%PT Crystal
Dielectric Constant	3900	8000
Dielectric Loss	0.025	0.008
Curie Temperature	210°C	166°C
Piezo. Coeff. d_{33} (pC/N)	690	2250
Coupling Constant k_{33}	0.79	0.91
Young's Mod. (GPa)	47	12
Mech. Quality Factor, Q_m	46	~50
Uses	Imaging, Linear and Phased Arrays	Experimental Imaging, Linear and Phased Arrays

electric performance; however, in the early '80's researchers at the Toshiba Corporation reported extraordinarily high coupling coefficients from (1-x)Pb($Zn_{1/3}Nb_{2/3}$)O_3-x$PbTiO_3$ (PZN-PT) relaxor ferroelectric single crystals [1]. Development of this material was slow, but in the mid '90's researchers at the Pennsylvania State University discovered that these crystals exhibited extremely high piezoelectric strain sparking a tremendous interest in using the material in a broad range of transducer applications [2]. To date large crystals (> 2.54 cm square) of both PZN-PT and especially PMN-PT ((1-x)Pb($Mg_{1/3}Nb_{2/3}$)O_3-x$PbTiO_3$) are readily available exhibiting electromechanical coupling coefficients in excess of 91% (see Table I). TRS Technologies, Inc. currently holds an exclusive license to Penn State's patents on the single crystal material for transducer and actuator applications [3,4] and is one of the world's primary suppliers of these materials.

Work on making ultrasound transducers from ferroelectric single crystals has clearly demonstrated that the high coupling coefficient does in fact result in very broad bandwidth. Research at Penn State on single element transducers made from 1-3 crystal-epoxy composites (Figure 2) with matching and backing layers has demonstrated bandwidths as high as 140% (Table II) [5]. In addition Toshiba has built crystal phased arrays with an 80% bandwidth [1] and Agilent has built crystal array transducers with a 95% bandwidth [6]. To date most of the work on crystal transducers has been done using conventional designs to compare crystal performance to that obtained with standard PZT. However, crystal production and uniformity are to the point that considerably more sophisticated arrays are now possible allowing the full exploitation of single crystals for harmonic imaging and high frequency applications.

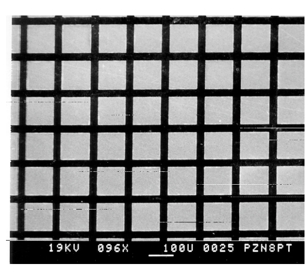

Figure 2: 1-3 Crystal-polymer composite constructed using dice-and-fill fabrication.

Table II: Bandwidths for a variety of single element crystal transducers fabricated from 1-3 crystal polymer composites. The bandwidth for lightly backed Type VI PZT transducer is in 70 to 80% range [5].

PZN-PT Vol%	Aperture (mm^2)	Matching Layer Impedance (MRayl)	Matching Layer Thickness (□)	Backing Impedance (MRayl)	Center Freq. (MHz)	Band Width (%)	Insertion Loss (dB)
69	30	3.3	0.31	0 (air)	4.1	74	-6.5
69	30	3.3	0.20	13.5	4.6	141	-18
50	36	6.5	0.30	6.0	3.5	97	-10

II. CRYSTAL GROWTH

PMN-PT single crystals are currently grown at TRS Technologies by the Bridgman method. This is a melt growth technique where starting materials (usually sintered PMN-PT ceramic) are placed in a sealed platinum crucible along with seed crystals and inserted into furnace with a sharp temperature gradient. The ceramic is melted but the seeds are positioned below the melt temperature in the furnace's temperature gradient. As the crucible is slowly pulled through the furnace the seeds promote formation of a single PMN-PT crystal in the melt. The growth runs are approximately 7-14 days in length. For harmonic imaging linear arrays it is expected that transducer sizes in excess of 20 x 20mm will be required. Crystals of this size are readily available using the Bridgman method as shown in Figure 3, a photograph of a PMN-PT crystal boule 4cm in diameter by >7cm long. Compared to Flux growth methods the larger sized crystals allow for simplified manufacturing techniques to be applied.

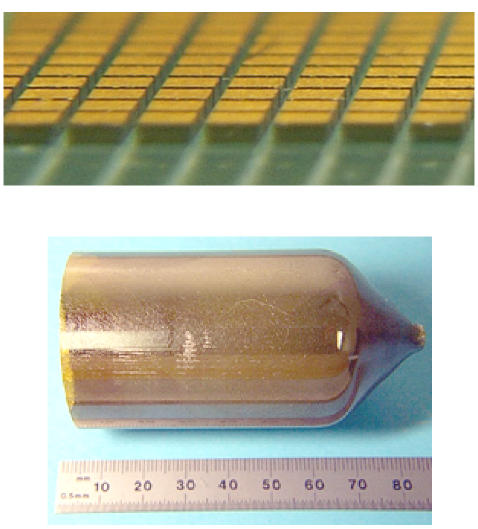

Figure 3: PMN-PT crystal boule (bottom) grown by the Bridgman method and a variety of crystal components produced from the boule by precision machining (top).

One problem with Bridgman grown PMN-PT is an axial composition gradient resulting from chemical segregation of the $PbTiO_3$ in the PMN-PT solid solution. The composition gradient results in piezoelectric property variation along the length of the boule. This in turn lowers the yield of high performance material per growth run and increases the cost. TRS Technologies is one of the few companies addressing this problem through use of the Zone Melting method. This method is shown schematically in Figure 4. In this technique only a portion of the ceramic starting material is melted at any one time limiting the amount of chemical segregation that takes place along the boule length by preventing convective flow in the melt. This results in a more compositionally uniform grown crystal. TRS has two 16-zone Bridgman furnaces for growing crystals by the zone-melting method. With this equipment the axial composition gradient can be kept to less that 0.4% per cm compared to nearly 1.4% per cm for conventional Bridgman growth methods as shown in Figure 5. The current growth capacity at TRS Technologies is four to six 4cm diameter boules per month.

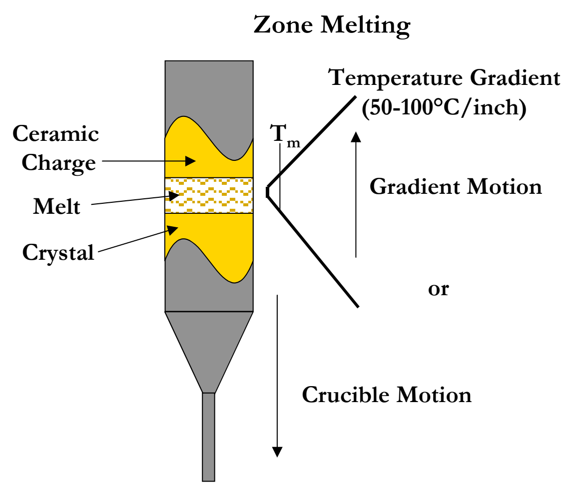

Figure 4: Schematic of Zone Melting growth configuration.

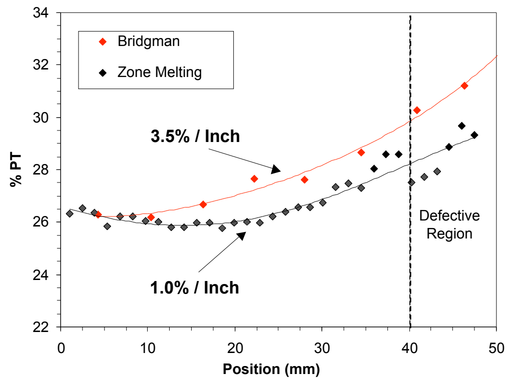

Figure 5. Compositional gradient comparison of PMN-PT single crystal grown by Bridgman and Zone Melting.

Single crystal piezoelectrics are clearly a revolutionary advance in piezoelectric technology. One of the factors that has limited their acceptance in the market place has been their very high cost compared to ceramics. Though crystals are still expensive, their cost has dropped dramatically since their initial development, and there is a strong impetus for costs to continue to drop due to demands of the medical ultrasound and Navy SONAR markets. TRS has been at the forefront of crystal cost reduction efforts by focusing on growth of PMN-PT by the Bridgman method. When first introduced, single crystal piezoelectrics were grown by the flux method. This approach yielded small irregularly shaped crystals after one month, resulting in expensive crystals. Because of this TRS has invested in Bridgman growth of PMN-PT. Because of this advance in crystal growth the price of single crystal PMN-PT has dropped from over $5/mm^3 to less than $0.8/mm^3, and we anticipate the price to drop further to $0.3/mm^3 by the end of 2003. Advances in the Zone Melting technique combined with scaling up the crystal diameter to >5cm, will directly contribute to the drop in price. The cost of single crystal is not likely to be as low as that of ceramic, but production can be scaled to bring the price of single crystal to within 2 to 3 times that of ceramic.

III. CONCLUSIONS

A broad variety of applications, including harmonic imaging and high frequency transducers, are currently under development. High electro-mechanical coupling make the piezoelectric single crystals very attractive for broadband ultrasonic and sonar transducers. Zone melting shows promise in allowing the growth of large uniform crystals required by these applications. As availability increases and cost decreases, single crystal piezoelectrics are expected to be an enabling technology in a broad range of actuator, sensor, and ultrasonic transducer applications.

IV. FUTURE WORK

Fully characterize broadband harmonic imaging transducers based on single crystal piezoelectrics and demonstrate their feasibility in enhancing resolution of ultrasonic images for specific organs or tumors. Develop and demonstrate single crystal piezoelectric composite transducers arrays with operating frequencies from 20-50Mhz using novel lithography based micromachining technology.

V. REFERENCES

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VI. ACKNOWLEDGEMENTS

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