

Innovations in Piezoelectric Materials for Ultrasound Transducers

Thomas R. Shrout

150 Materials Research Laboratory, The Pennsylvania State University, University Park, PA, 16802, USA

Abstract — Piezoelectric materials lie at the heart of ultrasonic transducers. For transducers used in medical imaging (3–7 MHz), PZT-5H ceramics offer high electromechanical coupling ($k_{33} \geq 75\%$), resulting in good bandwidth and sensitivity. As transducer arrays become smaller with increasing frequency, the development of high permittivity ($\epsilon_R^T > 7,000$ vs. 3,400 for PZT-5H), piezoelectrics based on polycrystalline PMN-PT, provide improved electrical impedance matching. Advanced medical diagnostic techniques, including contrast and harmonic imaging, have taken advantage of the recent development in single crystal Relaxor-PTs that offer coupling k_{33} 's $> 90\%$ and subsequently, significant increases in bandwidth. For small animal, ophthalmology and cellular imaging, higher resolution is demanded, thus requiring transducers operational in the range of 20–100 MHz. Advancements in ceramic processing include pore-free and fine-grain (≤ 1 micron) piezoelectric ceramics of PT and PZT, being an “enabling” technology, allowing the fabrication of high frequency single element and annular arrays. Innovations in the fabrication of high frequency arrays (≥ 30 MHz) include tape casting and sol-gel molding techniques. Of particular significance, DRIE (deep reaction ion etching), has demonstrated the ability to mill out ultrafine features, allowing 1–3 crystal-polymer composites operational at frequencies ≥ 60 MHz, far beyond that achieved by current state-of-the-art dicing.

INTRODUCTION

Innovations in piezoelectric materials are driven by the need for high performance transducers. Electromechanical coupling (k_{ij}), dielectric permittivity (ϵ_r) and acoustic impedance (Z) are the most important parameters which determine the performance of ultrasonic transducers [1,2]. Piezoelectric ceramics have been the material of choice, offering high coupling, and a range of permittivities. These merits translate into transducer performance in the form of high sensitivity and broad bandwidth. To efficiently couple the acoustic energy from high impedance piezoelectric ceramics ($Z \sim 30$ – 36 Mrayls) to the human body ($Z \sim 1.5$ Mrayls), quarter wavelength matching layers and/or low impedance composites comprised of piezoelectric ceramics with a passive polymer may be used. In addition, improved impedance matching composites offer geometries that utilize high coupling coefficient elements as depicted in Fig. 1.

The selection of the appropriate piezoelectric material is also based upon the frequency of operation.

Why Composites?

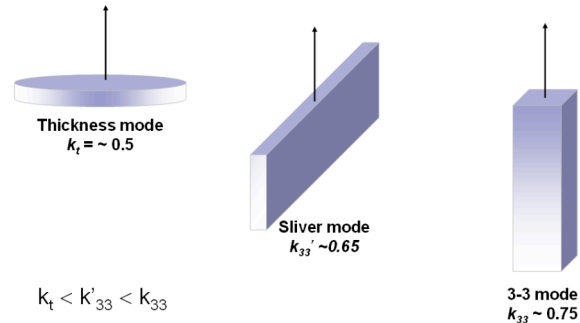


Fig. 1 Piezoelectric geometries and associated electromechanical coupling coefficients for single element transducers, 2-2 arrays and 1-3 composite transducers.

For ultrasonic imaging in the 3-7MHz regime, PZT-5H type ceramics have been the mainstay for array transducers offering electromechanical coupling $k_{33s} \geq 75\%$ and large dielectric permittivities ($\epsilon_r \geq 3400$). As transducer arrays become smaller with increasing frequency, piezoelectrics with higher permittivities are desired to maintain electrical impedance matching to the electronics. For very high frequency transducers $\gg 20$ MHz, as used in ophthalmic eye imaging, catheter-based intravascular imaging (IVUS) and small animal imaging, the ability to fabricate the associated fine scale features of the transducer elements becomes the critical issue.

In this review, recent developments in piezoelectrics as related to transducer requirements are presented including high permittivity ceramics, high coupling single crystals and novel fabrication methods.

PIEZOELECTRIC MATERIALS

The parameters of a few piezoelectric materials used in 1-3 composites and array transducers are presented in Table 1 and contrasted in figures 2 a & b. Developed more than 30 years ago, optimal PZT-5H ceramics possess a combination of high coupling and high permittivity while exhibiting a transition temperature $T_C \geq 200^\circ\text{C}$, which leads to improved temperature stability and minimal depolarization during fabrication. Furthermore, these materials are microstructurally pore free, a key aspect in transducer fabrication.

From figure 2a, higher permittivity piezoelectrics are readily available, but one must sacrifice T_C . Listed in Table I, PMN-PT and/or Relaxor-PZTs, e.g., $\text{Pb}(\text{Ni}_{0.5}\text{Nb}_{0.5})\text{O}_3$ based systems, offer permittivities nearly

twice that of the 5H materials, but exhibit $T_{CS} \leq 150^\circ\text{C}$. A fine grain version of the PMN-PT material (1-2 microns vs 5-10 microns) has been developed allowing ease of fabricating fine scale arrays by dicing [3]. Fine grain versions of PbTiO_3 (PT) have also been shown to be an enabling technology for the fabrication of 20-100MHz single elements and annular array transducers [4].

New MPB piezoelectrics in the $\text{Bi}(\text{Me})\text{O}_3\text{-PT}$ system have recently been found, offering good coupling, while exhibit $T_{CS} > 400^\circ\text{C}$, which have potential in transducers operational in harsh environments, e.g. NDE [5,6].

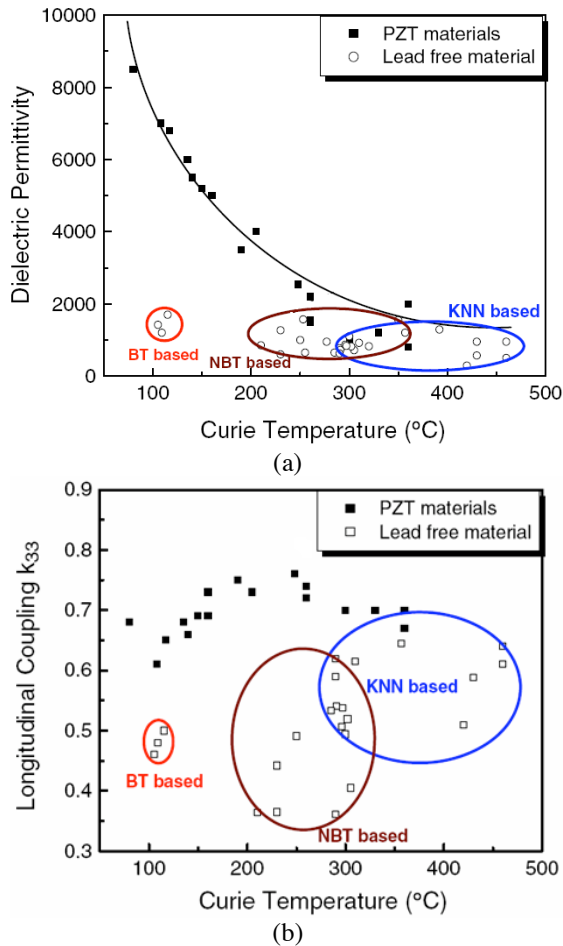


Fig. 2 Room temperature dielectric permittivity (a) and coupling coefficient (k_{33}) (b) as a function of Curie temperature for various PZT- based ceramics and lead free piezoceramics (BT-BaTiO₃, KNN-K,NaNbO₃, NBT-Na,BiTiO₃).

LEAD FREE PIEZOELECTRIC CERAMICS

With regulations of RoHS and WEEE to be enforced in Europe, enormous activities in the development of lead free ceramics to replace PZTs are ongoing. Shown in figures 2 a & b, data for various families of lead free piezoelectric ceramics clearly reflect that these materials exhibit far lower dielectric permittivities and electromechanical coupling than their lead based counterparts, thus limiting their usefulness in ultrasonic imaging to perhaps single element transducer(s) [8].

Table I. Properties of selected piezoelectric materials[7].

Property	PZT-5H	PMN-PT polycrystalline	PMN-32PT crystal
ϵ_r^T	3900	6000	8000
d_{33} (pC/N)	690	750	2250
k_{33}	0.80	0.74	0.91
N_3^T (Hz.m)	1300	1400	600
Curie Temperature ($^\circ\text{C}$)	210	150	166

Note: Piezoelectrics for high frequency (20-100MHz) single element transducers may include the low permittivity (≤ 100) materials PVDF, PbTiO_3 , LiNbO_3 and KNbO_3 crystals.

RELAXOR-PT SINGLE CRYSTALS

Advancements in medical diagnostic technologies including contrast and harmonic imaging, have taken advantage of the recent development of single crystal Relaxor-PTs that offer ultra high electromechanical coupling coefficients ($k_{33} \geq 90\%$) and thus high bandwidth. Advances in crystal growth and subsequent transducer fabrication have led to the commercialization of these novel crystals by Philips, Ibule and others, primarily ~5MHz cardiac imaging [9,10].

FABRICATION METHODS

An overview of various fabrication methods is given in figure 3 as a function of achievable scale and normalized acoustic frequency. The most common method used to fabricate 2-2 arrays and 1-3 composites is the “dice and fill” technique where a mechanical dicing saw is used to machine kerfs into a bulk piezoelectric and subsequently backfilled with an epoxy. The use of pore and defect free bulk materials insures the highest possible piezoelectric performance. The “dice and fill” technique, however, is limited in frequency when considering the small feature sizes of the piezoelectric elements and kerfs, the later to minimize unwanted lateral mode coupling.

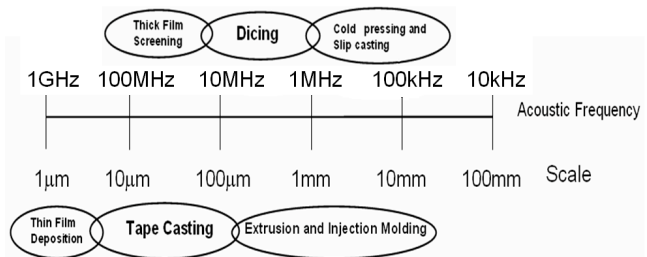


Fig. 3 Fabrication Methods and normalized frequency as a function of feature scale. Note: A frequency constant of $N_{33} = 1000\text{Hz.m}$ was assumed

For 2-2 composites, alternative methods to dice & fill have been used to create high frequency transducers (20-40MHz), including “tape-casting”, “stack and bond” and

“interdigital phase bonding”, however, these techniques are not amenable for high volume manufacturing [11-13]. As for 1-3 composites, the “lost mold” technique and related micro injection molding have been shown to produce composite transducers for frequencies >20MHz [14,15]. These techniques utilize powder processes and mixtures generally comprised of relatively large amounts of organic phase(s) which must be removed prior to densification. The subsequent polycrystalline ceramics are found to possess less than ideal microstructures and thus degraded performance, with coupling coefficients $k_{\text{eff}} \leq 40\%$.

Thin and thick film techniques include sputtering, chemical vapor deposition, sol-gel spin coating, inorganic/organic thick paste, etc., as well as polymer piezoelectrics such as PVDF, which offer features <10 microns and thus very high frequencies of operation. All these techniques suffer from low dielectric and electromechanical coupling ($k_t < 30\%$), due to clamping and defects as well as the inability to fabricate structures with high aspect ratios [16-18].

To overcome deficiencies of energetic and powder processes, micromachining of bulk materials has been recently developed to fabricate high frequency composite transducers (>40MHz). Figure 4 shows an SEM photo micrograph of a 1-3 composite PMN-PT single crystal prepared by deep reaction ion etching (DRIE). This repeatable and manufacturable process offers the transducers with coupling coefficient >70% [19-20].

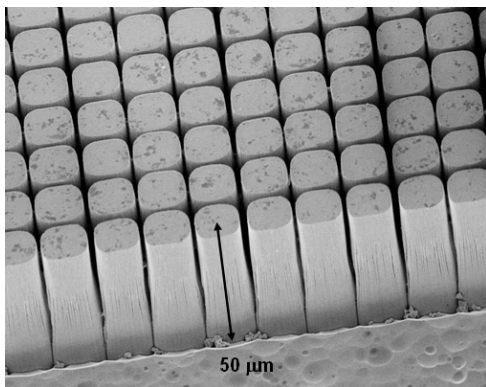


Fig. 4 SEM photo micrograph of a high frequency 1-3 single crystal composite fabricated by DRIE
Note- Kerf $\sim 5\mu\text{m}$

SUMMARY

Innovations in piezoelectric materials and fabrication methods for ultrasonic transducers have been primarily driven by the need for resolution in medical imaging. The development of Relaxor-PT single crystals with electromechanical coupling coefficients $k_{33} > 90\%$, significantly higher than current state of the art PZT-5H ceramic ($k_{33} \sim 75\%$), have made a significant impact, particularly in the area of cardiac harmonic imaging ($\sim 5\text{MHz}$). As transducer arrays become smaller with increasing frequency, the development of higher frequency (>20MHz) ultrasound transducers,” *Proc. Ultrason. Symp.*, pp.1253-1256, 2002.

permittivity ceramics, $\epsilon_{33} > 7000$ vs 3,400 for PZT-5H provides improved electrical impedance matching and thus performance. Advancements in ceramic processing including pore free and fine grain (≤ 2 microns) piezoelectrics, such as PT and PMN-PT, being an “enabling” technique for fabricating high frequency single element transducers and annular arrays

Innovations in fabricating high frequency 2-2 arrays and 1-3 composites include tape casting and sol-gel lost mold techniques. Of particular significance, DRIE (Deep reactive ion etching) has demonstrated the ability to mill out ultra fine features, allowing composites to be fabricated from bulk ceramics and single crystals for high frequency transducers.

Acknowledgement: The author would like to thank Drs. Shujun Zhang, Sorah Rhee, Xiaoning Jiang, and Jian Yuan for their input. The author also acknowledges the support of the Office of Naval Research (ONR) and National Institute of Health (NIH) under Grant #P41-RR11795.

References:

- [1] T. R. Gururaja, “Piezoelectrics for medical ultrasonic imaging,” *Am. Ceram. Soc. Bull.*, vol.73, pp.50-55, 1994.
- [2] W. A. Smith, “New opportunities in ultrasonic transducers emerging from innovations in piezoelectric materials,” *Proc. SPIE Int. Symp.*, vol.1733, pp.3-26, 1992.
- [3] H. Wang, B. Jiang, T.R. Shrout and W. Cao, “Electromechanical properties of fine grain 0.7PMN-0.3PT ceramics,” *IEEE Trans. Ultra. Ferro. Freq. Contr.*, vol.51, pp.908-911, 2004.
- [4] K.A. Snook, T. R. Shrout and K. K. Shung, “Design of a 50MHz annular array using fine-grain lead titanate,” *Proc. SPIE Int. Symp.*, vol.4687, pp.91-98, 2002.
- [5] R. E. Eitel, C. A. Randall, T. R. Shrout and S. E. Park, “Preparation and characterization of high temperature perovskite ferroelectrics in the solid-solution $(1-x)\text{BiScO}_3\text{-xPbTiO}_3$,” *Jpn. J. Appl. Phys.*, vol.41, pp.2099-2104, 2002.
- [6] S. J. Zhang, R. E. Eitel, C. A. Randall, T. R. Shrout and E. F. Alberta, “Manganese modified $\text{BiScO}_3\text{-PbTiO}_3$ piezoelectric ceramic for high temperature shear mode sensor” *Appl. Phys. Lett.*, vol.86, pp.262904, 2005.
- [7] www.trstechnologies.com
- [8] T. R. Shrout and S. J. Zhang, “Lead free piezoelectric ceramics: Alternatives for PZT?” *J. Electroceram.*, vol.19, pp.111-124, 2007.
- [9] S. E. Park and T. R. Shrout, “Characteristics of Relaxor-based piezoelectric single crystals for ultrasonic transducers,” *IEEE Trans. Ultra. Ferro. Freq. Contr.*, vol.44, pp.1140-1147, 1997.
- [10] US Patent: 6532819, “Wideband piezoelectric transducer for harmonic imaging,” 2003.
- [11] W. Hackenberger, S. Kwon, P. Rehrig, K. Snook and S. Rhee, “2-2 PZT-polymer composites for high
- [12] T.A. Ritter, T. R. Shrout, R. Tutwiler and K.K. Shung, “A 30-MHz piezo-composite ultrasound array

for medical imaging applications,” *IEEE Trans. Ultra. Ferro. Freq. Contr.*, vol.49, pp.217-230, 2002.

[13]R. Liu, K.A. Harasiewicz and F.S. Foster, “Interdigital pair bonding for high frequency ultrasonic composite transducers,” *IEEE Trans. Ultra. Ferro. Freq. Contr.*, vol.48, pp.299-306, 2001.

[14]Y. Hirata, K. Nakamae, T. Numazawa and H. Takada, “Piezoelectric composites of fine PZT rods realized by LIGA process,” *Sensors and Materials*, vol.16, pp.199-210, 2004.

[15]G. Pang, M. Sayer and G. R. Lockwood, “Fabrication of PZT sol gel composite ultrasonic transducers using batch fabrication micromolding,” *IEEE Trans. Ultra. Ferro. Freq. Contr.*, vol.53, pp.1679-1684, 2006.

[16]M. Lukacs, M. Sayer and S. Foster, “Single element high frequency (<50MHz) PZT sol gel composite ultrasound transducers,” *IEEE Trans. Ultra. Ferro. Freq. Contr.*, vol.47, pp.148-159, 2000.

[17] M. Sayer, M. Lukacs and T. Olding, “Emerging technologies for ferroelectric films and coatings,” *Integr. Ferroelect.*, vol.17, pp.1-10, 1997.

[18] Q.F. Zhou, K. K. Shung and Y. Huang, “Fabrication of sol-gel modified piezoelectric thick films for high frequency ultrasonic applications,” *Proc. Ultrason. Symp.* pp.1958-1961, 2004.

[19]J. R. Yuan, X. N. Jiang, P.J. Cao, A. Sadaka, R. Bautista, K. W. Snook and P. W. Rehrig, “High frequency piezo composites microfabricated ultrasound transducers for intravascular imaging,” *Proc. Ultrason. Symp.*, pp.264-267, 2006.

[20] X. N. Jiang, J. R. Yuan, A. Cheng, K. Snook, P. J. Cao, P.W. Rehrig, W.S. Hackenberger, G. Lavalle, X.C. Geng and T. R. Shrout, “Microfabrication of piezoelectric composite ultrasound transducers (PC-MUT),” *Proc. Ultrason. Symp.*, pp.918-921, 2006.