

Piezoelectric materials for high frequency medical imaging applications: A review

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Abstract The performance of transducers operating at high frequencies is greatly influenced by the properties of the piezoelectric materials used in their fabrication. Selection of an appropriate material for a transducer is based upon many factors, including material properties, transducer area, and frequency of operation. This review article outlines the major developments in the field of piezoelectrics with emphasis on materials suitable for the design of high frequency medical imaging ultrasonic transducers. Recent developments in the areas of fine grain and thin film ceramics, piezo-polymers, single crystal relaxor piezoelectrics, as well as lead-free and composite materials are discussed.

Keywords Piezoelectric · Thin films · Piezo-polymer · Composite materials · High frequency · Ultrasonic transducers

1 Introduction

Medical ultrasonic transducers come in a variety of forms and sizes ranging from single element transducers for mechanical scanning, linear arrays, to multidimensional arrays for electronic scanning. Although the performance of an ultrasonic scanner is critically dependent upon transducers/arrays, array/transducer performance has been one of the bottlenecks that prevent current ultrasonic imagers from reaching their theoretical resolution limit. The primary reason is that medical ultrasonic array/transducer design

and fabrication are processes of a broad interdisciplinary nature. They require knowledge from a variety of disciplines such as acoustics and vibration, electrical engineering, material sciences and engineering, medical imaging, and anatomy and physiology. It is of no wonder that even to date the design of transducers is still mostly empirical, generally involving a trial and error approach. Many manufacturers regard their expertise in designing and manufacturing transducers as a trade secret of the highest order.

The most crucial component of an ultrasonic transducer is a piezoelectric element. There are quite a number of piezoelectric materials available to choose from when designing a single element or array transducer. A critical look at the piezoelectric properties of these materials in the orientation of interest can greatly narrow down the choices. Materials such as single crystal lithium niobate (LiNbO_3), polycrystalline lead titanate (PbTiO_3), and polymer PVDF are excellent choices for fabricating large aperture single element transducers due to their exhibited low dielectric permittivities. In contrast, a material such as Navy Type VI (PZT-5H) is more ideal for array elements which typically have much smaller element areas due to its large dielectric permittivity.

High frequency ultrasonic imaging has many clinical applications because of its improved image resolution. It is gaining acceptance as a clinical tool for the examination of the anterior segment of the eye, skin and vascular system [1, 2]. Its development has pushed the limits of ultrasonic imaging technology, giving diagnostic quality information about microscopic structures in living tissue. For high frequency devices the grain size of the piezoelectric material does factor into manufacturability and attainable performance. Therefore in recent years quite a bit of research has been directed to the development of fine grain piezoelectric ceramics [3, 4], and single crystal piezoelectric materials

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[5–7]. This review article outlines the major developments in the field of piezoelectrics with emphasis on materials suitable for the design of high frequency medical imaging ultrasonic transducers.

2 Basic principles

Naturally occurring piezoelectric crystals are seldom used today as transducer materials in diagnostic ultrasonic imaging because of their weak piezoelectric properties. The most popular material is a polycrystalline ferroelectric ceramic material, lead zirconate titanate, $\text{Pb}(\text{Zr}, \text{Ti})\text{O}_3$ or PZT which possesses very strong piezoelectric properties following polarization. Polarization of a ferroelectric material is carried out by heating it to a temperature just above the Curie temperature of the material and then allowing it to cool slowly in the presence of a strong electric field, typically in the order of 20 kV/cm, applied in the direction in which the piezoelectric effect is required. The electrical field is usually applied to the material by means of two electrodes. This process aligns the dipoles along the direction of polarization. There are a great variety of ferroelectric materials. Barium titanate (BaTiO_3) was among the first that were developed. Certain piezoelectric properties of PZT can be enhanced by doping. As a result, many types of PZT are commercially available.

To completely describe the piezoelectric properties of a material, 18 piezoelectric stress constants and 18 receiving constants are required. Fortunately, since these materials usually are symmetric, a smaller number of constants are actually needed. For instance, in quartz there are only five constants. Unlike single crystals such as quartz and lithium niobate which have crystallographic axes, the principal axis of a ferroelectric ceramic like PZT is defined as the direction in which the material is polarized. A plate cut with its surface perpendicular to the x -axis of a crystal is called x -cut, and so forth. The x, y, z directions are indicated by numbers 1, 2, 3. For polarized ceramics, the 3 direction is usually used to denote the polarization direction. A piezoelectric strain constant, d_{33} , represents the strain produced in the 3 direction by applying an electric field in the 3 direction and d_{13} is the strain in the 1 direction produced by an electric field in the 3 direction when there is no external stress. Here it is important to note that the piezoelectric properties of a material depends upon boundary conditions and therefore upon the shape of the material. For example, the piezoelectric constant of a material in the plate form is different from that in a rod form.

The ability of a material to convert one form of energy into another is measured by its electromechanical coupling coefficient defined as stored mechanical energy/total stored energy. The total stored energy includes both mechanical

and electrical energy. It should be noted that this quantity is not the efficiency of the transducer. If the transducer is loss less, its efficiency is 100% but the electromechanical coupling coefficient is not necessarily 100% because some of the energy is stored as mechanical energy but the rest may be stored dielectrically in a form of electrical potential energy. Since only the stored mechanical energy is useful, the electromechanical coupling coefficient is a measure of the performance of a material as a transducer. The piezoelectric constants for a few important piezoelectric materials are listed in Table 1.

There are a few important material geometries that are frequently used in ultrasonic imaging. These are shown in Fig. 1 using the effect of geometry on electromechanical coupling coefficient as an example. For a disc of PZT-5H with the diameter much larger than the thickness (c), the thickness mode electromechanical coupling coefficient k_t is used. For a long tall element with the sides of the square cross-section much smaller than the thickness (a) and a tall element with rectangular cross-section (b), the electromechanical coupling coefficients are denoted respectively as k_{33} and k_{33}' . The geometry shown in Fig. 1b is the most important in that it is often used in linear arrays. It is interesting to note that the electromechanical coupling coefficient is highest in the bar mode shown in (a), attributable to the fact that more energy is irradiated in the long axis direction because the bar is surrounded by air which has a much lower acoustic impedance than PZT. The effect of material shape on other measured piezoelectric properties can be found in Table 2. If a 75 MHz transducer element, for example, is designed using PZT-5H it would be approximately 25–30 μm thick depending on the geometry used. However for array transducers the width of such an element, operating in bar or element mode, should be on the

Table 1 Properties of a few important piezoelectric materials used in high frequency transducer designs.

Property	PVDF [9, 10]	PZT-5H [8, 11]	PbTiO ₃ [11, 12]	PMN-PT Crystal (33%PT) [13, 14]
d_{33} (10^{-12} C/N)	-33	593	60	5,500–6,500
k_t	0.12–0.15	0.51	0.49	0.58
k_{33}	–	0.75	0.51	0.94
$\varepsilon_{33}^S/\varepsilon_0$	5–13	1,470	180	680–800
c (m/s)	2,200	4,580	5,200	4,610
ρ (kg/m^3)	1,780	7,500	7,660	8,060
Curie temp. ($^{\circ}\text{C}$)	100	200	260	130

$\varepsilon_{33}^S/\varepsilon_0$ is the relative clamped dielectric permittivity and ρ and c denote density and plate mode sound velocity, respectively. These values are approximate and will vary with the manufacturing and/or testing methodology used.

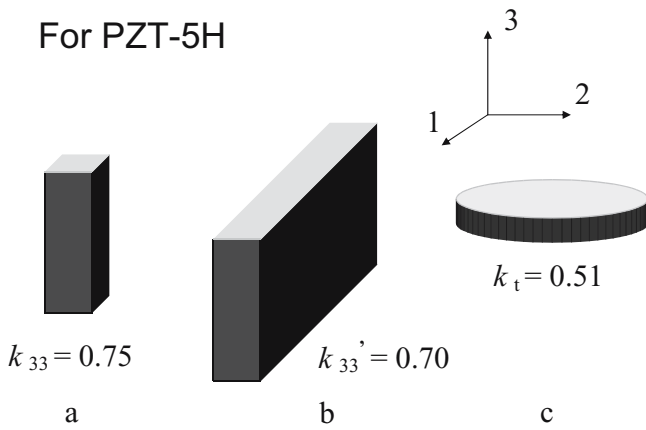


Fig. 1 The effect of a piezoelectric material affects its piezoelectric properties. This is illustrated by way of the electrical mechanical coupling coefficient **a** a tall element of square cross-section frequently seen in composite materials **b** a tall element frequently seen in linear arrays **c** a circular disc frequently seen in single element transducers [8]

order of 15 μm or less to avoid an unwanted lateral resonance. It is therefore easy to imagine how challenging it would be to fabricate such a device.

3 Fine grain ceramics

Conventional piezoceramics, having grain sizes on the order of 5 to 10 μm, are not particularly suited for high frequency array applications where the dimensions of the transducer element can be on the order of tens of micrometers. As mentioned in the pervious section if a PZT-5H element is used in a 75 MHz array transducer it would be on the order of 25 μm or 2.5–5 grains thick. Finer grain materials also have proven to be superior in dicing operations [3], and retain their bulk material properties better at higher frequencies than their larger grain counterparts [11, 16]. Lastly materials such as $Pb(Mg_{1/3}Nb_{2/3})O_3-PbTiO_3$ (PMN-PT), which are particularly suited for linear array transducers due to their high clamped dielectric permittivity, are now being manufactured in fine grain versions [17]. Alternately, fine grain $PbTiO_3$ has enabled the development of high frequency (50 MHz) annular array transducers due to its moderately low clamped dielectric permittivity [12].

4 Piezoelectric polymers

In addition to PZT, piezoelectric polymers have also been found to be useful in a number of imaging applications [18]. One of these polymers, polyvinylidene fluoride (PVDF), has been used for years to produce high frequency transducers [19, 20]. After processes like polymerization, stretching, and poling, a thin sheet of PVDF with a thickness on the order of 6 to 25 μm can be used as a transducer material. The advantages of this material are its low acoustic impedance

(4 Mrayls), flexibility, and cost. Transducers made from this material are typically very wideband. However this material is not very efficient due to its low transmitting constant, large dielectric loss and low electromechanical coupling coefficient ($k_t \sim 0.12-0.15$). Also it is not well suited for linear or phased array designs because of its extremely low relative clamped dielectric permittivity ($\epsilon_{33}^S/\epsilon_0 \sim 5-13$). Although PVDF is not an ideal transmitting material, it does possess a fairly high receiving constant. Miniature PVDF hydrophones are commercially available operating at up to 100 MHz. P(VDF-TrFE) co-polymer has been shown to have a higher electromechanical coupling coefficient ($k_t \sim 0.20-0.30$) [10] than PVDF and can be polarized by applying an electrical field at elevated temperatures. This material therefore has proven to be useful in the production of simple, yet very effective, high frequency single element and annular array transducers [21, 22].

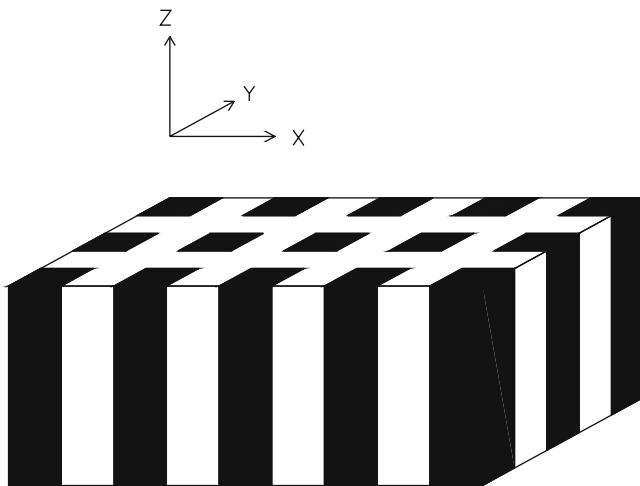
5 Piezoelectric composites

One of the most promising frontiers in transducer technology is the development of piezoelectric composite materials [23]. Innovation in fabrication technology has allowed the preparation of PZT polymer 1–3 composites for applications in the 1 to 80 MHz range [24, 25]. These 1–3 composites consisting of small PZT rods embedded in a low density polymer, illustrated in Fig. 2a where the dark and light regions indicate respectively the piezoelectric ceramic phase and the polymer phase, have been used for low-frequency underwater applications for many years. The notation 1–3, 2–2, etc has been coined by [26]. A notation of 1–3 means that one phase of the piezoelectric is connected only in one direction whereas the second phase in all three directions. A notation of 2–2 means that both phases are connected in two directions as illustrated in Fig. 2b. These composites with piezoelectric volume concentration of 20 to 70%, have a higher k_t , and lower acoustic impedance (4 to 25 MRayls) than conventional PZT (34 MRayls) which better matches the acoustic impedance of human tissue. The higher coupling coefficient and better impedance matching can lead to higher transducer sensitivity and improved image resolution.

Table 2 Approximate material properties for PZT-5H in different geometries [15].

Property	Bar mode	Element mode	Plate mode
Velocity (m/s)	3,850	4,000	4,580
Acoustic impedance (Mrayl)	28.9	30.0	34.4
Electromechanical coupling coefficient	0.75	0.70	0.51

a 1-3 Composite



b 2-2 Composite

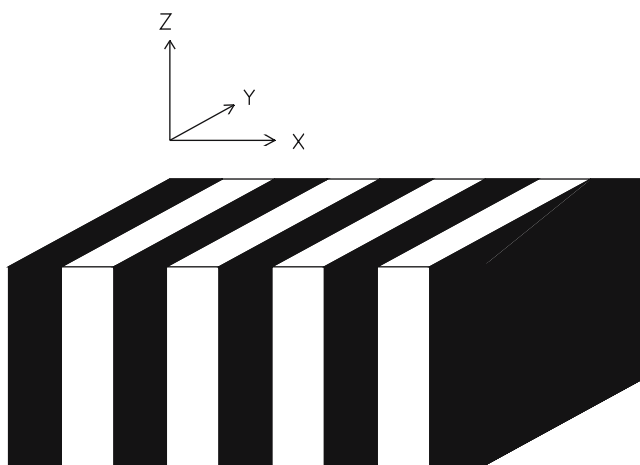


Fig. 2 Two different configurations of piezoceramic composites **a** 1–3 composites **b** 2–2 composites [26]

There exist a number of techniques currently available for fabrication of piezo-polymer composites. The most common of these is the “dice-and-fill” technique whereby a mechanical dicing saw is used to cut kerfs into a piece of bulk piezoceramic, which are subsequently backfilled with epoxy [27]. Unfortunately this technique can be problematic when considering the small feature sizes required for very high frequency transducer operation. Interdigital pair bonding [24] and more recently interdigital phase bonding [25] have been determined to be effective modified dicing methods for manufacturing high volume fraction 2–2 or 1–3 composites at frequencies up to 80 MHz. Other composite machining techniques available include laser ablation, and

chemical etching which are more useful when working with brittle single crystal materials at high frequencies. The stacking and bonding of thin piezoceramic plates has also been an effective method for creating high frequency 2–2 composites [28]. An electron micrograph of a 2–2 composite manufactured by this technique is shown in Fig. 3 with 18 μm wide piezoceramic posts and 5 μm wide polymer kerfs. Using tape-cast PZT is a viable alternative to this “stack-and-bond” technique for production of large quantities of 2–2 composites at low cost [29]. This technique involves the printing of carbon black ink on thin sheets of green piezoceramic tape. The tape layers are then stacked to form the 2–2 structure, and the carbon is volatilized during heat treatment of the stack. The voids left by the removed carbon can then be back-filled with epoxy under vacuum. Another technique that is suitable for large scale production is termed the “lost-mold” technique [30]. This technique involves the pressing of a piezoceramic paste into mold, which is usually silicon and can be designed for various composite geometries. The mold is removed by chemical etching. After sintering the remaining composite posts, the voids left by the mold removal are backfilled with epoxy. It should be noted however that at present both the tape-cast and lost-mold techniques exhibit inferior piezoelectric properties when compared to composites fabricated using bulk piezoceramic or single crystal materials.

6 Relaxor-based piezoelectric single crystals

For several decades, the material of choice for medical transducers has been polycrystalline ceramics based on the solid solution $\text{Pb}(\text{Zr}_{1-x}\text{Ti}_x)\text{O}_3$ (PZT), compositionally engineered near the morphotropic phase boundary (MPB). The search for alternative MPB systems has led researchers to revisit relaxor-based materials with the general formula, Pb

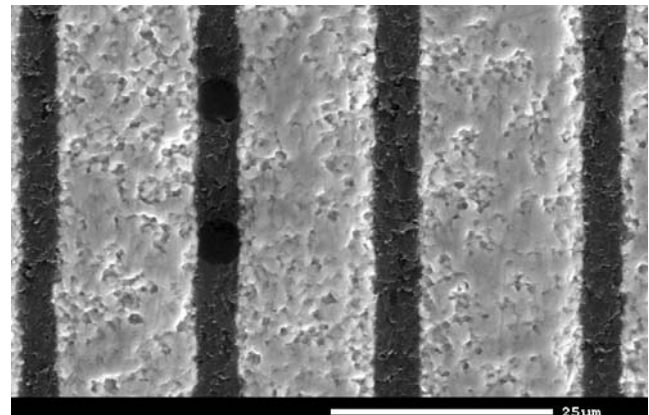


Fig. 3 A scanning electron micrograph of a 2–2 composite consisting of a piezoceramic separated by polymer [28]

$(B_1, B_2)O_3$ ($B_1: Zn^{2+}, Mg^{2+}, Sc^{3+}, Ni^{2+} \dots, B_2: Nb^{5+}, Ta^{5+} \dots$). In the single crystal form, however, Relaxor-PT materials, represented by $Pb(Zn_{1/3}Nb_{2/3})O_3-PbTiO_3$ (PZN–PT), $Pb(Mg_{1/3}Nb_{2/3})O_3-PbTiO_3$ (PMN–PT) have been found to exhibit high coupling coefficients ($k_{33} > 0.9$), dielectric constants ranging from 1,000 to 5,000 with low dielectric loss $< 1\%$, and exceptional piezoelectric coefficients $d_{33} > 2,000$ pC/N [5]. However, these single crystals have low Curie temperatures ($T_c < 160$ °C) which may limit their thermal stability during operation in medical imaging devices. Thus, there is currently an interest in exploring materials with similar piezoelectric properties and higher Curie temperatures (> 300 °C). It has been reported that single crystal $Pb(Yb_{1/2}Nb_{1/2})O_3-PbTiO_3$ (PYbN–PT) can have a Curie temperature as high as 360 °C and a d_{33} as high as 2,500 pC/N [31, 32]. This material therefore may 1 day replace PMN–PT and PZN–PT as the material of choice for medical imaging transducers.

Several groups have used relaxor-based piezoelectric single crystals for medical ultrasonic transducer designs [33]. Saitoh et al. [34] reported a 3.7 MHz phased array transducer using PZN-9%PT single crystal. This array displayed increased sensitivity (5 dB) and bandwidth (25%) over the equivalent PZT array. Oakley and Zipparo [7] reported on a 4 MHz 1–3 PZN–PT single crystal composite transducer with 100% –6 dB bandwidth and a 6 MHz PMN–PT transducer with 114% bandwidth. Ritter et al. [6] reported –6 dB bandwidths of 75–141% for 3.5–4.5 MHz single element PZN–PT composite transducers. Subsequent to these works, single crystal transducers for cardiac harmonic imaging applications have been commercially realized at approximately 5 MHz by Philips Ultrasound [33]. However, further investigation in crystal growth, micro-machining techniques and testing are still required if these materials are to be used effectively in high frequency applications.

7 Lead free materials

As previously mentioned the most widely used piezoelectric ceramics are lead oxide based ferroelectrics, especially

Table 3 Relevant material properties of 36° Y-cut $LiNbO_3$ and 49.5° X-cut $KNbO_3$.

Property	$LiNbO_3$ [37]	$KNbO_3$ [38]
k_t	0.49	0.67
$\epsilon_{33}^S / \epsilon_0$	39	45
ρ (kg/m ³)	4,640	4,570
c (m/s)	7,340	7,600
Acoustic impedance (Mrayl)	34.0	34.7
Curie temp. (°C)	1,150	220

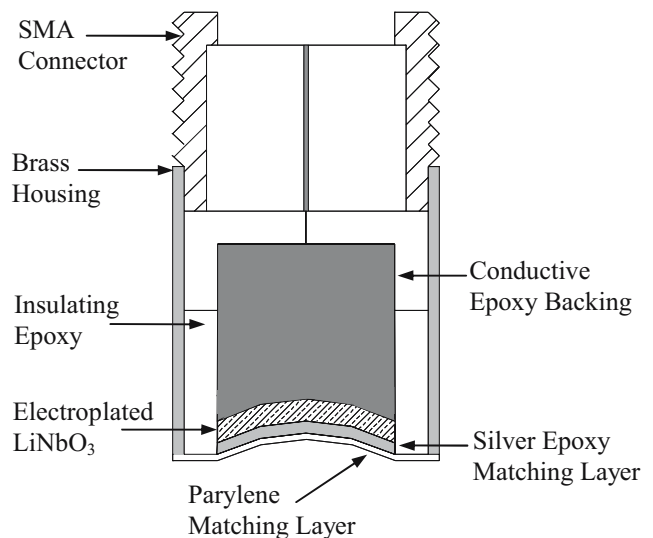


Fig. 4 Design cross section of a typical high frequency $LiNbO_3$ transducer. This drawing is not to scale

the $Pb(Zr,Ti)O_3$ (PZT) system. However, the PZT materials contain up to 50% lead by weight. Due to the high toxicity of lead, there is significant interest in developing lead-free piezoelectric ceramics and single crystals for biomedical transducer applications. Recently, Saito et al. [35] reported a new combination of a morphotropic phase boundary in an alkaline niobate-based perovskite solid solution, and the development of a processing route leading to highly (001) textured polycrystals in the $(K, Na)NbO_3-LiTaO_3$ system and $(K, Na)NbO_3-LiTaO_3-LiSbO_3$ system. The ceramic developed exhibits a d_{33} of above 300 pC/N, and the texturing material leads to a peak d_{33} of 416 pC/N. Hollenstein et al. [36], also reported that potassium sodium niobate piezoelectric ceramics substituted with lithium $(K_{0.5-x/2}, Na_{0.5-x/2}, Li_x)NbO_3$ or lithium and tantalum $(K_{0.5-x/2}, Na_{0.5-x/2}, Li_x)(Nb_{1-y}, Ta_y)O_3$ can be synthesized by traditional solid state sintering. The compositions chosen are among those recently reported to show improved piezoelectric properties. However these materials do not currently perform as well as their lead containing counterparts and have yet to prove their worth in medical imaging transducer designs.

Single crystal lithium niobate ($LiNbO_3$) and potassium niobate ($KNbO_3$) are lead free materials that have proven to possess several material properties ideal for making single element transducers especially at high frequencies. These materials display a comparable k_t to PZT, a low dielectric permittivity and high longitudinal sound speed, which are all ideal for designing sensitive large aperture high frequency single element transducers (Table 3). Numerous transducers operating the 10–100 MHz range have been constructed using both of these materials [37–39]. The performance of these devices varied due to design differences with –6 dB bandwidths up to 75% and insertion loss

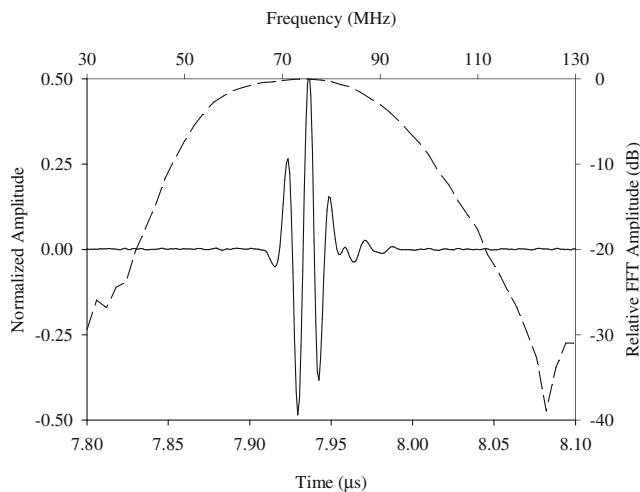


Fig. 5 Measured pulse-echo response for a 75 MHz LiNbO_3 transducer. The axes on the *top* and *right* refer to the pulse frequency spectrum (*dashed line*)

values as low as 9 dB. Figure 4 shows the design cross-section and Fig. 5 shows the echo waveform and spectrum for a representative 75 MHz high frequency LiNbO_3 transducer. Results from these studies have shown that LiNbO_3 and KNbO_3 are two of the most ideal materials available for fabricating highly sensitive single element transducers for high frequency applications. In fact the high sensitivity of transducers constructed using these materials has enabled clinical images to be attained at higher frequencies than what was possible with other materials used in the past [40, 41].

8 Piezoelectric thin films

One of the technical challenges for very high frequency (>100 MHz) imaging is the fabrication of transducers with piezoelectric elements of a thickness on the order of only a few micrometers. It is very difficult and time consuming to lap down and mechanically machine very thin piezoelectric

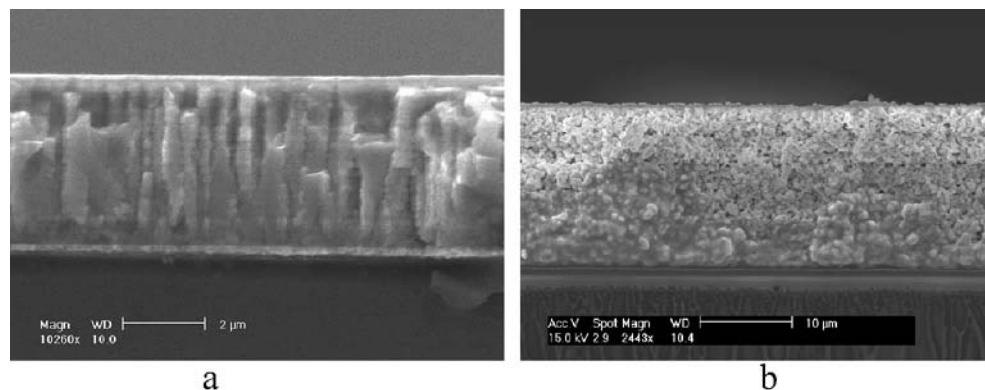
ceramics and crystals. Therefore, piezoelectric thin/thick film technology may be a good low-cost alternative solution for high frequency transducer (100–200 MHz) designs. However, the only drawback to using these materials is their inferior piezoelectric properties when compared to bulk produced piezoceramics [42–48].

To date, there are a number of groups that have successfully fabricated PZT thin/thick films. Sugiyama et al. [42] prepared PLZT thick films by a multiple electro-phoretic deposition and sintering processes. Barrow et al. [43] and Lukacs et al. [44] have reportedly used thick PZT ceramic coatings using sol-gel derived porous 0–3 composites to produce high frequency single element transducers. Chen et al. [45] produced PZT thick films by a modified sol-gel process involving an acetic acid route. Kurchania and Milne [46] also fabricated PZT thick films using titanium di-isopropoxide bi-acetylacetonate as a precursor material. Finally, Zhou et al. reported the development of PZT composite films [47], and the fabrication of micro-electromechanical (MEMS) tonpilz transducers using modified sol-gel PZT films [48]. The scanning electron microscope (SEM) cross-section of a 6 μm and 20 μm thick sol-gel film are shown in Fig. 6a and b, respectively. It is anticipated that films such as these will be used to fabricate >100 MHz array transducers in the near future.

9 Summary and conclusions

In this article we reviewed the current state of art in piezoelectric materials for high frequency applications. Fine-grain ceramics are easier to machine and have proven to retain their bulk material properties better at higher frequencies than their larger grain counterparts. These materials are currently the ideal choice for high frequency linear and phased array designs. Piezoelectric polymers are widely used in high frequency single element transducer designs because of their low acoustic impedance, inherently wideband nature, flexibility, and low cost. Piezoelectric composites have proven to be superior in performance

Fig. 6 A scanning electron micrograph of PZT films deposited on silicon substrate derived by the **a** pure sol-gel process and **b** modified process



when compared to traditional bulk piezoelectrics and piezoelectric polymers. However these materials are difficult to fabricate at high frequencies due to dimensional requirements. Nevertheless there exist several novel composite manufacturing techniques available for transducers designed to operate at up to 80 MHz. Relaxor-based single crystal materials have superior electromechanical coupling over traditional piezoelectric ceramics but have yet to be proven effective in high frequency transducer designs. Lead free materials provide an alternative to traditional lead based piezoelectric materials in designs where biological safety is an issue. Some common lead free materials such single crystal lithium niobate (LiNbO₃) and potassium niobate (KNbO₃) have proven to be ideal for fabricating highly sensitive, large aperture, high frequency single element transducers. Finally, thin-film deposition and micromachining techniques are an exciting new area of study for the development of very high frequency (>100 MHz) single element transducers and arrays.

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