# PIEZOELECTRIC MATERIALS FOR ULTRASONIC TRANSDUCERS : REVIEW OF RECENT DEVELOPMENTS

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#### ABSTRACT

First, a brief review of available classes of piezoelectric materials (in particular emergent piezoelectric single crystals) and specific requirements for medical imaging applications is made. Several piezoelectric materials are retained for two specific applications (50 MHz high frequency single element transducer and high density 2D array) to simulate and compare the overall transducer performance. This is performed by combining the K.L.M model to calculate the parameters and an optimisation method to extract the best design which gives the transducer performance. Single crystals are shown to be promising, particular for high density arrays.

# INTRODUCTION

Transducer performance requirements are more and more demanding due to a wide range of imaging modalities, such as flow and harmonic modes, which are now integrated in echographic systems. Transducer performance depends on its internal structure and on constitutive material properties, the most important of which is the active piezoelectric element. During the past fifty years, a succession of new piezoelectric material as been discovered, from Barium titanate (BaTiO3) in the 1950s to piezoelectric single crystal based on a relaxor (PZN or PMN) and PT solid solution, constantly improving the electromechanical properties [1]. This paper gives a brief review of the piezoelectric materials and pertinent parameters used for medical imaging applications.

To study and quantify, the overall performance of a transducer integrating these different piezoelectric materials, it is very difficult to define a universal figure of merit for a large range of configurations or applications. So, two precise configurations have been retained. First a high frequency single element transducer with a centre frequency at 50 MHz will be simulated with six different piezoelectric materials. Many applications require such transducers as dermatology, ophthalmology or small animal imaging. Secondly, one element of a high density 2D array with a centre frequency at 3.5 MHz is studied. These results will allow to show which piezoelectric material delivers the highest transducer performance for a precise configuration.

# **GENERAL AND SPECIFIC REQUIREMENTS**

The classical single-element transducer, in particular for medical imaging applications, is based on a piezoelectric plate or disc poled along the thickness direction, whose thickness

defines the resonance frequency of the device. The plate, typically a ferroelectric ceramic, has an acoustic impedance (*i.e.* around 33 MRa) much higher than of biological tissues (close to that of water, *i.e.* 1.5 MRa). This large difference leads to an acoustical mismatch and a poor axial resolution. Consequently, other layers are added to the active layer. On the front (*i.e.* between the piezoceramic and the propagation medium), matching layers are used. The thickness of a matching layer is generally around a quarter-wavelength at the resonance frequency, and its acoustical impedance is intermediate between those of the piezoceramic and tissues. The use of a matching layer thus improves the sensitivity of the transducer. Moreover, since the acoustical energy can better flow towards the tissues, the duration of acoustical resonance in the active layer is decreased. Consequently, the matching layer also improves axial resolution. The use of multiple matching layers, based on the same principle, can further improve transducer performance.

On the rear face of the active element, a thick layer is usually added. It is referred to as the backing which allows acoustic energy to flow by the rear face. The closer its acoustical impedance is to that of the active layer, the more energy is lost. The consequence is a lower sensitivity but a higher axial resolution. Thus, a trade-off has to be performed for each application. The attenuation coefficient and the thickness of the backing layer must be sufficient so that no energy can be radiated back to the active layer, which would produce parasitic echoes.

For the piezoelectric material, in particular for medical applications, two of the most important material parameters for transducer applications are the effective electromechanical coupling coefficient  $k_{eff}$  of the main vibration mode used and the acoustic impedance Z.

The  $k_{eff}$  factor represents the piezoelectric activity of the material in the considered mode of vibration. It should be as high as possible. This factor depends not only on the material properties but also on the geometry of the active element. In medical imaging applications, all vibration modes are longitudinal, *i.e.* the displacements are in the poling direction which defines the thickness dimension. For large plates or discs (thickness much lower than lateral dimensions), the thickness coupling factor  $k_i$  is used. For bars or pillars (thickness higher than lateral dimensions), the factor is  $k_{03}$ . For the intermediate case of an array element (one small and one large lateral dimensions with a thickness value between them),  $k'_{33}$  factor is defined. The value of the dielectric constant also has an important role on the electrical matching.

# COMMONLY USED PIEZOELECTRIC ELEMENTS

Figure 1 represents the values of the thickness mode coupling factor ( $k_t$ ) versus the acoustic impedance (Z) for a wide range of available piezoelectric materials. It can be observed that no material allows to obtain both high coupling and acoustic impedance equal to that of biological tissues. For almost all medical transducer applications, PZT piezoceramics are used because of their high coupling factor, even through their acoustic impedance is high, which can be compensated by using acoustic matching layer in the transducer structures. Many types of PZT piezoceramics are available on the market, with different properties due to doping by additives in varying proportions and to specific fabrication processes. For a given application, properties such as dielectric constant and grain size allow to choose a specific material. A wide range of references can be found from relatively low (a few hundred) to very high (a few thousand) relative dielectric constants with grain sizes from one to ten micrometers. For large area devices such as single element transducers, a moderate dielectric constant allows good electrical matching to cables and electronics (which are typically at 50 to 80 ohms), while array elements require much higher dielectric constants. For high frequency devices or for arrays (whenever small dimensions must be achieved), fine grain materials are required.

Considering these requirements, soft PZT materials are commonly used such as PZT-5A (or Ferroperm Pz27) with moderate dielectric constant or PZT-5H (or Ferroperm Pz29) with higher dielectric constant.

At the end of the 70's, piezocomposites combining a high coupling piezoceramic (such as soft PZT) and a low acoustic impedance polymer (such as an epoxy resin) appeared. These materials allow to obtain a very good trade-off between a high coupling and a low acoustic impedance. For ultrasonic medical transducer applications, ceramic pillars in a polymer matrix (called 13 connectivity) are of great interest. A high thickness coupling factor  $k_t$ , close to the value of  $k_{03}$  in the ceramic alone, is obtained even for relatively low ceramic contents, for which the acoustic impedance is around three to four times lower than that of pure ceramic. These

properties allow both sensitivity and bandwidth to be increased in transducers, in comparison with classical piezoceramic devices.



Figure 1 : Electromechanical coupling factor in thickness mode  $(k_t)$  versus the acoustical impedance for different piezoelectric materials.

## **NEW MATERIALS AND TECHNOLOGIES - FUTURE TRENDS**

#### Materials

#### New piezoceramic compositions

Piezoceramic manufacturers are constantly improving the properties of their materials, so new references often appear on the market. The trends in the past few years have been on one hand to increase dielectric constant while maintaining high coupling factors, and on the other hand to decrease grain size and porosity. This is achieved by developing ceramics based on PZT but with relatively large proportions of additives, but also materials based on a solid solution between PZT phase and a relaxor phase leading to compositions such as PLZT or PNNZT, and by optimising fabrication processes. Ferroperm Pz21 is an example of this type of high performance piezoceramic [2].

#### High coupling single crystals

At the beginning of the 80's, publications on the PZN-PT solid solution system have reported very high coupling coefficients, higher than those of standard ceramics if the appropriate axis is utilised (namely  $k_{33}$  around 90%). Studies such as those by Shrout et *al.* [3] have investigated properties of several families of single crystals and in 1994, Toshiba in Japan patented such single crystals for ultrasonic transducer applications, in particular medical imaging. Currently, two families are being investigated: PZN-PT and PMN-PT. Piezoelectric single crystal growth, generally by the Bridgman process [4], takes several weeks to obtain large size with high quality. Today, the largest PZN-PT single crystals made with this process are 80 mm diameter (the mass is higher than 1kg) and 50 mm diameter for PMN-PT composition [4]. The maximum operating temperatures are lower than those of standard PZTs but are generally sufficient for medical applications. Different lead titanate quantities can be used in the compositions. The properties are modified, in particular the value of the permittivity which decreases for an increasing lead titanate proportion. This allows a choice of compositions as a function of applications and transducer design. In the same time, electromechanical properties (in particular  $k_{33}$ ) are relatively stable.

Longitudinal wave velocity  $(v_{33})$  is lower but density is slightly higher as compared to soft PZT, resulting in a significantly lower acoustic impedance. The very high values of  $k'_{33}$  and  $k_{33}$  are of interest for 22 connectivity (classical linear array technology) and 13 connectivity piezocomposites. However, using the dice and fill technique, risks of breakage and chipping in the single crystal material are increased and repoling is necessary to obtain the expected electromechanical performance.

Lead-free single crystals have also been developed, namely KnbO<sub>3</sub> which displays  $k_{33}$  values close to those of PZN-PT and values of  $k_t$  similar or higher than those of both PZN-PT and PMN-PT compositions. Such compositions are very promising for single element transducers, in particular at high frequencies.

Several transducer applications of single crystals have been published [5]. Single element transducers using 13 composites operating around 4 MHz [6] have shown both high sensitivities and large bandwidths. In array configuration, an increase of sensitivity of 5 dB and a 25% larger bandwidth have been obtained in comparison with a PZT-5H (type 600) based array. Finally, B-mode images of the left ventricle of an adult heart [7] have been produced using a 128 element PZN-PT array operating at 3.5 MHz.

#### Simulations

To compare precisely the capability of the available materials, two configurations have been chosen here. First, for high frequency application, a small piezoelectric disk is integrated in a single element transducer and secondly materials will be used to simulate one element of a high density array. All these simulations will be performed with the K.L.M. model (equivalent electrical circuit) coupled with an optimisation method devoted to medical imaging where the propagation medium is considered as water. This method is based on a minimisation of a performance index which contains three parameters calculated with the impulse response of transducers (two related to the axial resolution and one to the sensitivity). Details on this method is given in [8].

# Thick film or fhin Plate for high frequency applications

Six different piezoelectric materials have been retained for comparison. Piezoelectric copolymers are used for two reasons: low acoustic impedance close to that of water and flexibility which allows an easy focusing with no lens. The LiNbO<sub>3</sub> crystal and lead titanate ceramic have a high thickness coupling factor and a low dielectric constant. The PZT soft ceramic will be used as reference material and finally new single crystals (two compositions PMN-33%PT and PZN-8%PT) will be compared.

For all these simulations, two parameters have been fixed: the transducer centre frequency (50 MHz) and the active area of the piezoelectric element (diameter of 3 mm *i.e.* an area of  $7.07 \text{ mm}^2$ ).

In the simulations, a self inductor (in serial), a transformer and a 50 Ohm cable of 1.5 m is also taken into account. Table 1 gives all the parameters of piezoelectric materials used for the simulations.

Material	k <sub>t</sub> (%)	ε <sub>33</sub> <sup>S</sup> /ε <sub>0</sub>	ρ (kg/m <sup>3</sup> )	v₁ (m/s)	δ <sub>e</sub> (%)	δ <sub>m</sub> (%)	Z (MRa)
P(VDF-TrFE) [9]	29	4.1	1932	2380	6.9	4.0	4.6
LiNbO <sub>3</sub> crystal [10]	49	28	4640	7340	0.1	0.01	34.1
PT ceramic [10]	49	200	6900	5200	0.9	0.83	35.9
PZT soft ceramic [11]	50	800	7900	4390	2.5	2.7	34.7
PMN-33%PT [12]	63	680	8060	4645	-	-	37.4
PZN-8%PT [6]	48	560	8200	4120	-	-	33.8

Table 1 : Piezoelectric material parameters for high frequency single element transducers (k<sub>t</sub>: thickness coupling factor;  $\epsilon_{33}^{S}/\epsilon_{0}$ : clamped dielectric constant;  $\rho$ : density;  $\gamma$ : longitudinal wave velocity;  $\delta_{e}$ : dielectric losses; ;  $\delta_{m}$ : mechanical losses; Z: acoustic impedance).

The optimisation of the performance of each transducer allows to determine the characteristics of one or two matching layers (acoustical impedance and thickness), of the backing (only the acoustical impedance since this medium is supposed semi-infinite), the value of the self inductor and the turns ratio of the transformer. All these results are specified in Table 2 with the values of the insertion loss calculated as the ratio between the received power and the power delivered by the generator in pulse echo mode and the bandwidth at -6 dB.

These simulations do not take into account the possibility of focusing by addition of lens. These results allow, for an identical configuration, to quantify the gain and compare the piezoelectric materials. Impulse responses are represented on Figure 2. Only the transducer based on the P(VDF-TrFE) has been simulated with no matching layer, since the acoustical matching will be not of great interest for the final performance. Due to its relatively low k, the sensitivity is low in comparison with all other simulations. Two matching layers are used for the other transducers. The results for the PbTiO<sub>3</sub> and LiNbO<sub>3</sub> are similar. The electrical matching allows to compensate for the difference in the dielectric constants. The PZT soft ceramic delivers a similar bandwidth but a lower sensitivity. Finally, the single crystal (PMN-PT) gives the best trade-off with the highest sensitivity and bandwidth (except for the copolymer). The PZN-PT also gives good results (not mentioned in Table 2) but slightly lower than those of PMN-PT.

Material	е	Zb	Z <sub>m1</sub>	e <sub>m1</sub>	Z <sub>m2</sub>	e <sub>m2</sub>	tuning	BW	IL
	(µm)	(MRa)	(MRa)	(×λ/4)	(MRa)	(×λ/4)		(%)	(dB)
P(VDF-TrFE)	22.4	3.3	-	-	-	-	Yes	74	27
LiNbO <sub>3</sub> crystal	69.6	6.6	7.1	0.78	2.1	0.8	Yes	55	17.4
PT ceramic	51.0	3.1	5.2	0.82	1.8	0.9	Yes	51	16.6
PZT soft ceramic	43.1	6.7	4.6	0.71	1.7	0.7	Yes	55	19.3
PMN-33%PT	44.3	1.5	4.9	0.75	1.9	0.8	Yes	62	16.8

Table 2 : Design and performance parameters of high frequency single element transducers (e: thickness of the piezoelectric material;  $\zeta_{2}$ : acoustic impedance of the backing;  $Z_{n1}$  and  $Z_{m2}$ ; acoustic impedances of the first and second matching layers;  $e_{n1}$  and  $e_{n2}$ : thicknesses of the first and second matching layers;  $e_{n1}$  and  $e_{n2}$ : thicknesses of the first and second matching layers; BW: bandwidth at –6dB; IL: Insertion loss).

## High density 2D arrays

Five materials are used for simulations of a 2D array element (Table 3). The size of the element is  $250\mu$ m×250 $\mu$ m and the centre frequency is fixed at 3.5 MHZ. In this case, k<sub>33</sub> is the coupling factor, v<sub>33</sub> and Z<sub>33</sub> the corresponding wave velocity and acoustical impedance. Two PNNZT ceramics are used with slightly different dielectric constants and acoustical impedances. The PZT soft ceramic is kept as reference and a single crystal composition (PMN-33%PT) is also tested.

Material	k <sub>33</sub> (%)	$\epsilon_{33}^{S}/\epsilon_{0}$	ρ (kg/m <sup>3</sup> )	v <sub>33</sub> (m/s)	δ <sub>e</sub> (%)	δ <sub>m</sub> (%)	Z <sub>33</sub> (MRa)
Ferroperm Pz21 [2]	71	1800	7780	3700	2.5	1.1	28.8
PNNZT [11]	72	2130	8150	4210	3.5	1.4	34.4
PZT soft ceramic [11]	66	800	7900	4240	2.5	2.7	33.5
PMN-33%PT [12]	94	680	8060	3340	-	-	27.0

Table 3 : Piezoelectric material parameters for high density arrays ( $k_{33}$ : coupling factor;  $\epsilon_{33}$ <sup>S</sup>/ $\epsilon_0$ : clamped dielectric constant;  $\rho$ : density;  $v_{33}$ : longitudinal wave velocity;  $\delta_e$ : dielectric losses; ;  $\delta_m$ : mechanical losses;  $Z_{33}$ : acoustic impedance).

The results are summarised in Table 4. The PZT soft ceramic has the lowest performance due to relatively low  $k_{33}$  and dielectric constant. The two PNNZT ceramics give similar performance. The single crystal delivers very high performance with an increase of more than 40% for the bandwidth and a gain around 2dB dB for the sensitivity. Figure 3 shows also the impulse responses.



Figure 2 : Impulse responses of the two simulations (a- single element transducer, b one element of a 2D array) for a 60 V amplitude and 20ns duration pulse.

Material	е	Z <sub>b</sub>	Z <sub>I1</sub>	e <sub>l1</sub>	Z <sub>l2</sub>	e <sub>l2</sub>	tuning	BW	IL
	(μm)	(MRa)	(MRa)	(×λ/4)	(MRa)	(×λ/4)		(%)	(dB)
Ferroperm Pz21	29.8	2.1	3.4	1	1.7	1	No	59	16.4
PNNZT	33.9	1.2	3.7	1	1.7	1	No	58	15.8
PZT soft ceramic	39.9	1.0	4.4	1	1.8	1	No	47	16.8
PMN-33%PT	19.7	2.3	4.1	0.9	1.8	0.8	No	106	14

Table 4 : Design parameters and performance of high density arrays.

# CONCLUSION

Different piezoelectric materials have been compared in two configurations which do not require the same properties, in particular in term of dielectric constant. However, the results show that, in both cases, single crystals allow to obtain the best transducer performance. For 2D array, a gain of 2 dB for the sensitivity and 40% for the bandwidth is achieved in comparison with PNNZT.

To this day however, these single crystals are not commercially available in large size, with reproducible and homogeneous properties and at affordable prices. High permittivity ceramic compositions such as PNNZT are still the best choice for an industrial production, namely for high density arrays.

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