PROPERTIES OF COMPOSITE PIEZOELECTRIC MATERIALS FOR ULTRASONIC TRANSDUCERS

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ABSTRACT

A new spectrum of electromechanical properties are realized with composite materials made by combining a piezoelectric ceramic and a passive epoxy. We have studied disks of rod composites in which the epoxy resin holds together parallel thin rods of piezoelectric ceramic oriented perpendicular to the faces. If the lateral spatial periodicity of the structure is smaller than all the relevant acoustic wavelengths, these composites behave as a homogeneous piezoelectric with new "effective" material parameters. For ultrasonic transducer applications we can engineer the structure to make materials whose electromechanical coupling is higher and acoustic impedance is lower than those of conventional piezoelectric ceramics.

1. Introduction

Conventional piezoelectric ceramics, such as lead zirconate-titanate (PZT), lead metaniobate and modified lead titanates, are useful for making ultrasonic transducers used in medical imaging because of their high electromechanical coupling coefficients (k_t ~0.4 - 0.5). Compositions with dielectric constants ranging from 200 to 4800 provide the flexibility to adjust the material to the system electronics. Their principal limitation lies in their high acoustic impedance (~20 - 30 Mrayls) which makes coupling to tissue (~1.5 Mrayls) difficult. Acoustic matching layer technology has been developed to address the need for efficient, broadband coupling to tissue required to produce the compact pulses used in pulse-echo medical imaging.

Piezoelectric polymers, such as polyvinylidene difluoride (PVDF) and its copolymers [1] present a different set of material properties. Their low acoustic impedance (~4 Mrayls) simplifies efficient broadband coupling, but their low electromechanical coupling ($k_t \leq 0.3$), low dielectric constant (~10) and high dielectric losses ($\tan\delta$ ~15%) present additional limitations when integrating these materials into medical imaging transducers.

The emergence in recent years of composite piezoelectric materials [2-4] opens up the possibility of engineering new materials with electromechanical properties better suited to medical ultrasound transducers. Such composite materials are built up by combining a piezoelectric ceramic with a passive polymer. A great variety of structures have been made; each offers new options in material properties. The class of structures we report on here is shown schematically in Figure 1. These rod composites, or more formally 1-3 PZT-polymer composites, consist of parallel rods of piezoelectric ceramic held together by an epoxy matrix. Such structures offer improved electromechanical properties and the possibility to tailor those properties to optimize system performance.

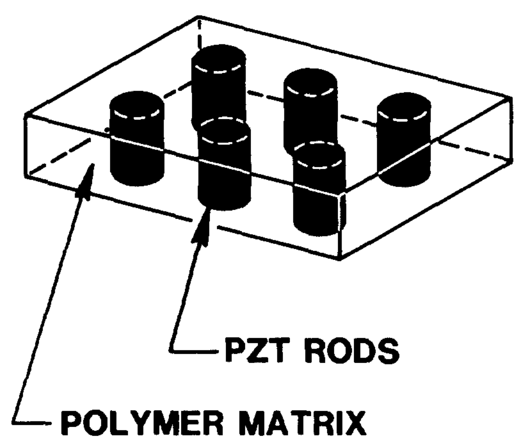


Figure 1. Schematic representation of 1-3 PZT-polymer, or rod, composites consisting of PZT rods in a polymer matrix [After Newnham et al.[2-4]].

In this paper we report on the physical considerations behind designing composite piezoelectric materials for use in thickness mode transducers. The following paper reports on the performance obtained from single-element and linear-array transducers made from these composite piezoelectrics.

2. Material Fabrication

A number of techniques have been developed to fabricate rod composites both at Pennsylvania State University [4,5] and within our own lab [6]. Figure 2 illustrates one method based on dicing part-way through a solid disk of PZT with a multiple-blade semiconductor wafering saw. A polymer epoxy is then poured into the grooves. After curing the epoxy, the base of solid PZT is cut away leaving a thin disk consisting of rectangular rods held together by the polymer similar to the structure shown schematically in Figure 1.

The measurements reported in this paper were made on disks fabricated in this fashion from PZT5A [7] and Spurr epoxy [8]. These are the same materials used to make the samples studied at Pennsylvania State and Stanford Universities that are reported in the three preceding papers [9-11]; so direct comparisons can be made. The spatial periodicity of these samples is about 500 microns with 250 micron square rods, i.e. approximately 25% PZT by volume.

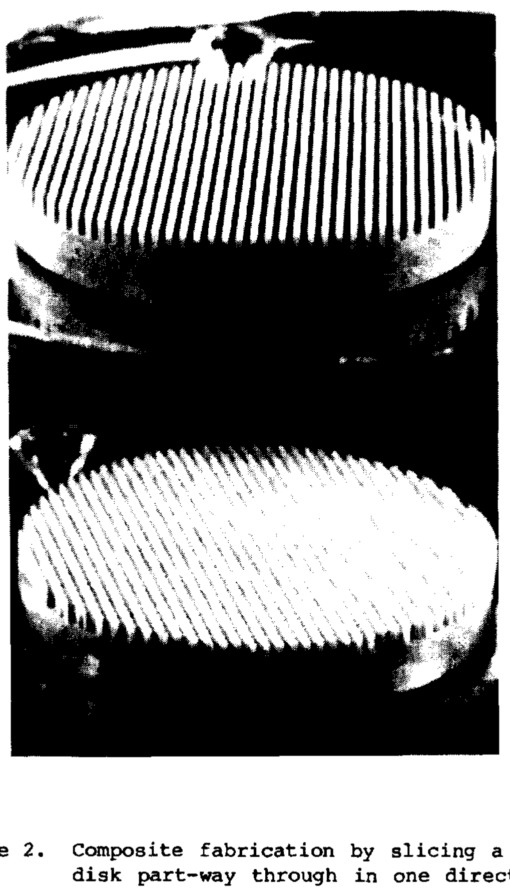


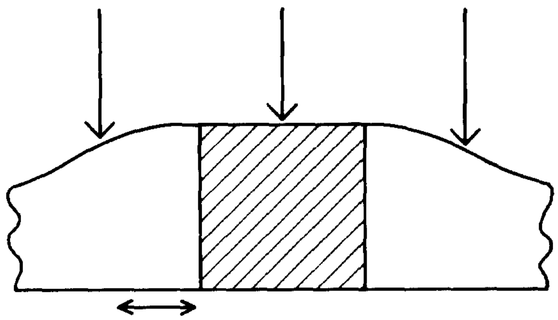
Figure 2. Composite fabrication by slicing a PZT disk part-way through in one direction and again in the perpendicular direction.

3. Theory

Let us first take up the question of when we can consider a rod composite structure to behave as a homogeneous material with new "effective" material constants. The preceding papers [9,10] present a detailed study of the very complex modes in rod composite disks due to stop-band resonances in the lateral periodic array of rods. For a given periodicity, there is a whole spectrum of resonant Lamb waves on a composite plate, beginning at the frequency of the first stop-band resonance. It would be a catastrophe to have these lateral resonances occur near the thickness-resonance mode of the disk that we wish to use as an ultrasonic transducer in that thickness mode. We can avoid this regime by making composite plates with a sufficiently fine lateral spatial scale, compared with the thickness, that its lateral stop-band resonances occur at frequencies well above the fundamental thickness mode of the plate. Viewed in the spatial domain, such a choice insures that all excited acoustic waves have wavelengths large compared with the lateral variations in composition. That is, of course, if we limit the excitation frequencies to be near the thickness mode frequency. In such a case the acoustic waves "average" over the unresolved fine structure much as the acoustic waves in the usual piezoelectric ceramic "average" over properties of individual ceramic grains. Thus the material can be considered an effective homogeneous piezoelectric with "new" properties if the lateral periodicity is sufficiently fine compared with its thickness.

We can get further physical insight into this requirement by considering the gedanken experiment shown schematically in Figure 3. Here a high-frequency pressure wave is impinging on a single PZT rod embedded in a compliant polymer plate. The polymer is soft both under compression and shear so its surface distorts dramatically near the stiffer PZT rod. The ceramic rod thus stiffens the structure in its vicinity. This stiffening extends out some fraction of the epoxy's shear wavelength at the driving frequency. Now if the polymer plate is filled with a periodic array of PZT rods whose spacing is much smaller than this shear wavelength, the polymer is effectively "tied" to the rods. The plate then oscillates uniformly with the ceramic and polymer moving together. Thus the pressure exerted on the epoxy is not lost but effectively transferred to the PZT rods which produce the electric response.

Considering the case where the rod composite plate oscillates uniformly near its thickness-mode resonance we can readily calculate the new "effective" material parameters describing that resonance. The easiest properties to determine are the density and dielectric constant of the composite. They are, in first order, just the average of these properties of the component phases weighted by volume fraction. For the density this is clear because it is a scalar quantity; for the dielectric permittivity it follows from simple summation of parallel capacitances.



$$d \sim \frac{1}{n}\lambda_{shear} = \frac{1}{n}\frac{v_{shear}}{f}$$

$$d \sim \frac{v_{shear}/n}{f}$$

Figure 3. Schematic representation of a high-frequency pressure wave impinging on a single PZT rod in a polymer plate. For a distance, d, from the rod the polymer is effectively tied to the ceramic.

The elastic constant governing the thickness distortions is also just the volume-fraction weighted average of that of the component phases. This is readily seen using a parallel-springs picture much like the parallel capacitance viewpoint used to calculate the dielectric constant. Here however a judicious choice can improve the calculation with little effort. A compression of the plate will cause both the ceramic and the polymer to want to bulge in the lateral direction (i.e. the Poisson ratio effect). Since we will operate the transducer at frequencies high compared with lateral modes of the sample as a whole, the total lateral distortion is clamped or "frozen out". The component phases, however, have lateral scales corresponding to much higher frequencies than the thickness resonance so the substructure is not laterally clamped in detail. The ceramic and polymer will compete in the lateral direction, and the much stiffer ceramic will win. Indeed, the polymer will only exert a modest degree of clamping on the ceramic rod over its behavior as a totally free rod. Thus, in first approximation, it is best to use the elastic constant describing oscillations in a laterally free PZT rod, not a laterally clamped PZT disk. For the polymer it makes little difference, since its contribution to the elastic constant of the composite is small even in first approximation.

While the calculation of the density is exact, there are clearly corrections to the dielectric and elastic constants available in a more detailed theory. Flux leakage and relaxation of the lateral clamping on the rod give countervailing corrections to the ceramic's contribution to the dielectric constant. The partial lateral clamping of the rod by the polymer and the actual squeezing of the polymer by the ceramic, both serve to stiffen the composite over the first-order calculation.

From the values for the density and elastic constant, the longitudinal velocity, $(c/\rho)^{1/2}$, and specific acoustic impedance, $(\rho c)^{1/2}$, are readily calculated. For the velocity, the lower density and elastic constant cancel each other out and only modest change is observed from PZT behavior. For the specific acoustic impedance, the effects compound and dramatically lower values (~ 4 Mrayls) can be attained. The low acoustic impedance is an important advantage for rod composite piezoelectrics.

The effective electromechanical coupling coefficient of the composite disk receives no direct contribution from the polymer phase, since it is not piezoelectric. In first order we can take k_t for the composite to be just that of a free PZT rod (i.e. k_{33} for the ceramic). The partial lateral clamping of the polymer will serve to reduce the effective k_t somewhat, but nevertheless it will be still significantly larger than the fully lateral-clamped value (i.e. k_t) for the ceramic. By using a ceramic with high k_{33} significant enhancement can be obtained. The larger electromechanical coupling coefficient is another advantage for rod composite piezoelectrics.

4. Experiment

For the samples reported in this paper let us first note that the first-order theory is expected to be valid. The shear velocity of the Spurr epoxy (~ 1000 m/sec) yields a shear wavelength of 2 mm near the 500 kHz thickness resonance. This is much greater than the 250 micron spacing between the rods.

Table 1 shows the electromechanical properties of 25% PZT rod composite material along with those of its component phases. A full discussion of the measurements is presented below. But let us here note that the composite's effective properties are in good agreement with predictions of the first-order theory.

The remainder of this section describes, for one particular sample, a detailed experimental verification that rod composites can be accurately described as a homogeneous piezoelectric material with new "effective" material constants. The performance of a thickness-mode piezoelectric transducer can be accurately predicted [12] using the one-dimensional model embodied in the Mason [13] or KLM [14] equivalent circuits. Besides the transducer geometry, four material parameters are needed to predict the performance from a homogeneous piezoelectric material: the clamped dielectric constant, ε^S, the longitudinal acoustic velocity, v^D, the thickness electromechanical coupling constant, k_t, and the specific acoustic impedance, Z. Three of these (ε^S, k_t, v^D) are obtained directly by measuring the frequency

Table 1. Electromechanical properties of a typical 25% PZT rod composite and its component materials together with the predictions of a first-order theory. The underlined quantities are the directly measured ones. The others are inferred.

	PZT DISK	PZT ROD	SPURR	25% PZT COMPOSITE EXP	25% PZT COMPOSITE CALC
ε^T	1200	1200	4	280	300
$\rho(10^3\ kg/m^3)$	7.5	7.5	1.1	2.5	2.7
$c(10^{10}\ N/m^3)$	15	10.8	0.5	2.9	3.1
$Z(10^6\ rayls)$	34	28.5	2.4	8.5	8.4
$v(10^3\ m/sec)$	4.5	3.8	2.2	3.4	3.4
k(%)	46	65	0	60	65

dependence of the electrical impedance in the vicinity of the thickness-mode resonance in a thin disk sample using either the IRE standard [15] or the impedance circle technique [16]. The acoustic impedance, Z, is also determined directly by measuring the same resonance curve but with one face of the sample in water. The losses caused by acoustic radiation into water dominate, so we obtain Z by fitting the electrical impedance to its theoretical form,

$$\frac{1}{j\omega C_o}\left[1-\frac{k_t^2}{k\ell}\,\frac{2\tan(k\ell/2)-j(Zo/Z)}{1-j(Zo/Z)\cot(k\ell)}\right],$$

where $C_o = \varepsilon_o\varepsilon^S A/\ell$ is the clamped capacitance of the sample (area A, thickness ℓ), $k = \omega/v^D$ is the propagation constant of the acoustic wave and Z_o is the specific acoustic impedance of water.

We have performed these measurements using an HP4192A Impedance Analyzer controlled by an HP9825T Desktop Computer. *Figures 4 and 5* show the measured resonance curves and the theoretical fit for a sample with $k_t = 0.60$, $Z = 7.5$ Mrayls, $\varepsilon^S = 200$, and $v^D = 3400$ m/sec.

It is important to confirm that these materials parameters accurately describe the acoustic transduction in composite piezoelectric materials. To do this we have measured the reception sensitivity, transmission efficiency, and acoustic radiation profile from an air-backed 19 mm disk of material. Absolute pressure measurements were made using a calibrated 1 mm

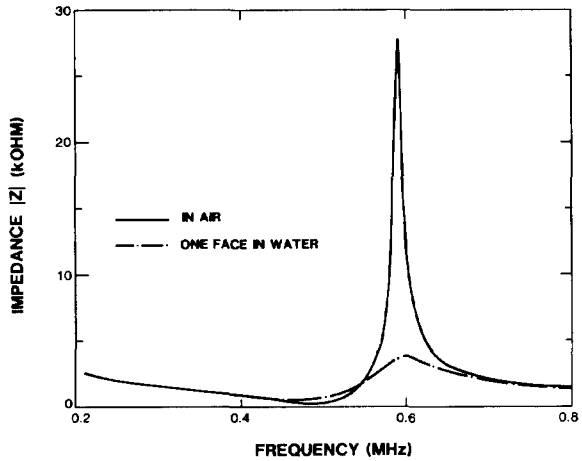


Figure 4. Magnitude of the electrical impedance measured near the thickness resonance of a composite disk both in air and with one face in water.

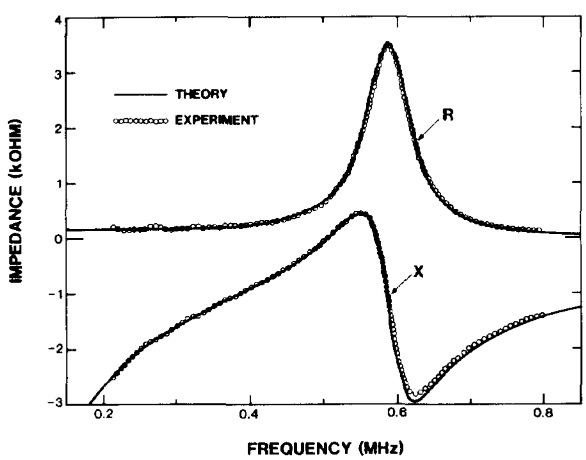


Figure 5. Real and imaginary parts of the electric impedance of a composite disk (with one face in water) measured near its thickness resonance plus the fitted theoretical curve which determines the material parameters.

diameter PVDF hydrophone probe [17,18]. Transmission efficiency is measured as the ratio of the pressure amplitude at a point in the far field along the disk's axis to the voltage amplitude across the sample. In measuring the reception sensitivity, a broadband unfocused transducer was used as a source with the sample perpendicular to its axis, well into the far field. The reception sensitivity was determined as the ratio of the open circuit voltage amplitude to the incoming pressure amplitude; minor corrections due to amplitude and phase variations of the pressure over the disk receiver were taken into account.

Figure 6 shows the transmission efficiency measured 12 cm along the axis of the same 19 mm disk whose materials constants were determined from the impedance measurements shown above. Figure 7 shows the reception sensitivity of this same sample. In both figures the theoretical fits to these absolute measurements were made using materials parameters that agree with those derived from the resonance fits; there are no other adjustable parameters. The agreement with theory is excellent. The largest source of experimental uncertainty is the ± 2.5 dB absolute calibration of the hydrophone probe below 1MHz [18] which influences only k_t; the other parameters agree well within 5%.

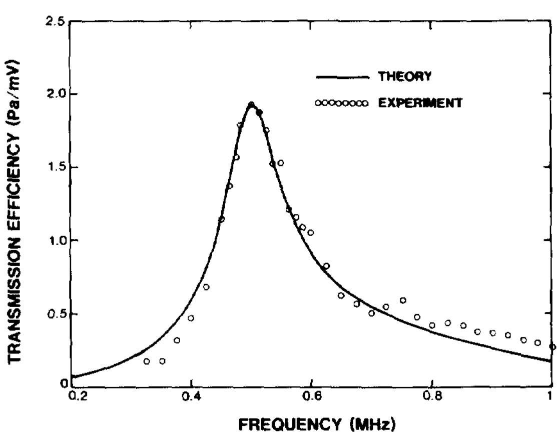


Figure 6. Absolute transmission efficiency measured 12 cm along the axis of a 19 mm diameter disk sample, together with the theoretical fit.

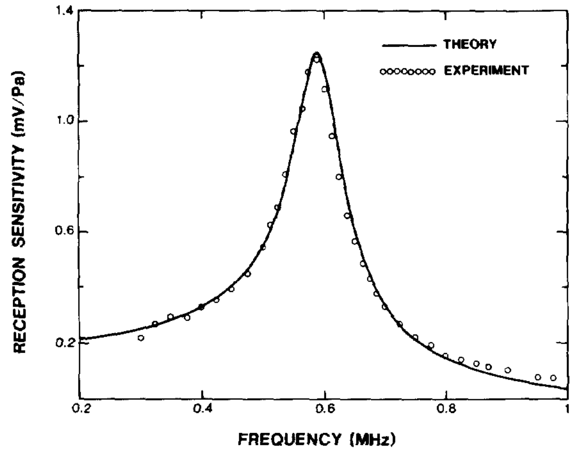


Figure 7. Absolute reception sensitivity measured in the far field of the source (corrected for minor phase and amplitude variations), together with the theoretical fit.

Figure 8 shows the acoustic radiation profile measured with a 1 mm diameter PZT hydrophone probe [DAPCO NP10-1] at 490 kHz in the far field. The theoretical curve is the beam profile expected for a 19 mm diameter flat-piston radiator. Again the agreement with theory is excellent.

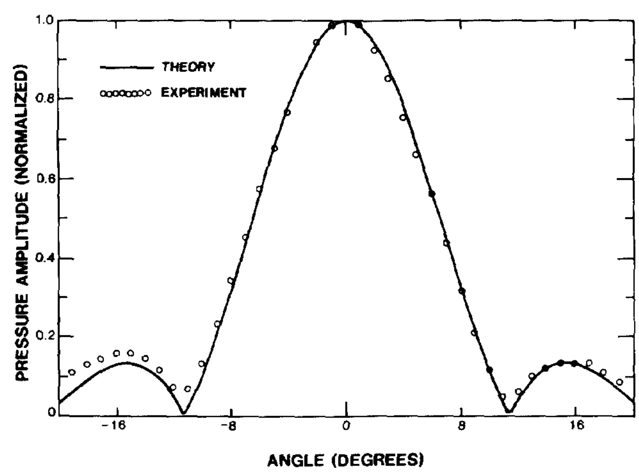


Figure 8. Acoustic radiation profile measured in the far field of a 19 mm disk sample at 490 kHz.

5. Conclusions

For thickness-mode ultrasonic transducers, a rod composite piezoelectric can be described as a homogeneous material with new "effective" electromechanical properties if the lateral spatial periodicity is sufficiently fine compared with the thickness. Making fine spatial scale material is the key to designing composite for use in thickness mode transducers at high frequencies.

Rod composite piezoelectrics have been made which have higher electromechanical coupling coefficient ($k_t \geq 0.6$) and lower acoustic impedance (≤ 7.5 Mrayls) than conventional piezoelectric ceramics. Such a piezoelectric material is particularly useful for making ultrasonic transducers for medical imaging.

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7. References

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