Lead Zirconate Titanate/Poly(vinylidene Fluoride-Trifluoroethylene) 1-3 Composites for Ultrasonic Transducer Applications

Kin Wing Kwok, Helen Lai Wah Chan, and Chung Loong Choy

***Abstract*—A new procedure for preparing lead zirconate titanate (PZT)/poly(vinylidene fluoride-trifluoroethylene) (P(VDF-TrFE)) 1-3 composites with both phases piezoelectrically active is described. Sintered PZT rods are inserted into a prepoled copolymer matrix, and the composite is repoled under a lower electric field. Using this new procedure, the dipoles in the two phases are aligned in either the same or opposite directions. Composite disks, of 12.7-cm diameter and 0.33- to 0.60-mm thicknesses, have been fabricated with PZT rods of 0.8 or 1 mm diameter distributed in a square pattern with 3 mm center-to-center separation. The ceramic volume contents of the composite disks are 3.6 and 5.6%, respectively. The resonance characteristics of the composite disks consist of the resonance modes of the two constituent phases, but they are dominated by the coupled longitudinal thickness mode (H-mode) of the PZT rods. The coupled radial mode (L-mode) resonance of the PZT rods is significant only for thin disks. The observed resonance frequencies of the H- and L-modes agree well with the values calculated from the coupling theory. The thickness mode resonance of the copolymer matrix (T-mode) is present but hardly observable in thick disks. The composite disks have been fabricated into transducers with air-backing and with no front face matching layer, and their performance characteristics have been evaluated in water. The transmitting and receiving voltage responses of a PZT/P(VDF-TrFE) composite transducer are better than those of a PZT/epoxy composite transducer. The transmitting and receiving voltage responses are improved when the PZT rods and copolymer matrix are poled in opposite directions, especially when the resonance frequencies of the H- and T-modes are approximately equal. When the phases are poled in the same direction and the resonance peaks associated with the H- and T-modes just overlap, the bandwidth is improved. Using 0.33-mm thick composite disks, a transducer can be produced with three operating frequencies by poling the constituent phases in the same direction, or with two operating frequencies at equal efficiency by poling the constituent phases in opposite directions. The PZT/P(VDF-TrFE) 1-3 composite transducer, especially the one with multiple operating frequencies, should be very promising in the applications of medical ultrasonic imaging.**

I. INTRODUCTION

PIEZOELECTRIC ceramic/polymer 1-3 composites have been used widely in medical ultrasonic imaging because of their relatively high electromechanical coupling coefficient (k_t) and low acoustic impedance (Z_o) as compared with the constituent ceramic [1]–[6]. Smith *et al.* [1] found that lead zirconate titanate (PZT)/epoxy 1-3 composite with 25 volume percent of PZT-5A has a k_t value larger than that of the PZT 5A ceramic (0.6 vs. 0.46) and a Z_o value closer to that of human tissue (8.5 vs. 35 Mrayl). A variety of composites have been fabricated using different constituent materials and different methods, and their properties and performance as transducers have been studied in detail.

Onoe and Tiersten [7] had extended the coupling theory developed by Giebe and Blechschmidt [8] for purely elastic vibrations to predict the coupled resonance frequencies of a piezoelectric vibrator of finite size. The calculated resonance frequency as a function of the width-to-thickness ratio for a PZT-5 rectangular plate agreed with the experimental data obtained [7]. Chan and Unsworth [5] and Chan *et al.* [6] studied the coupled resonance characteristics of square ceramic rods in ceramic/epoxy 1-3 composites. The calculated resonance frequencies agreed well with the observed values for PZT-5H/epoxy, PZT-7A/epoxy 1-3 composites [5] and for modified lead titanate/epoxy 1-3 composite [6]. In addition, they showed that, for medical imaging applications (operating frequency > 5 MHz), the width-to-length ratio of the ceramic rods should be larger than 1.5 for composites having a 0.075-mm polymer width and a 0.15-mm ceramic width [6]. Bui *et al.* [9] followed the work of Chan and Unsworth [5] to fabricate a dual-frequency (2 and 5 MHz) ultrasonic transducer using PZT-7A/epoxy 1-3 composites.

However, most of the 1-3 composites are made of piezoelectrically active ceramic and piezoelectrically passive polymer, and relatively little information is available on the properties of 1-3 composites with both phases piezoelectrically active. Because piezoelectricity can be activated in bulk, unstretched copolymer of vinylidene fluoride-trifluoroethylene (P(VDF-TrFE)), it can be used as the matrix for the composites. Taunaumang *et al.* [10] prepared PZT/P(VDF-TrFE) 1-3 composites using the dice-and-fill and lamination techniques, but these approaches resulted in alignment of the dipoles of the PZT phase only. Because the coercive field of P(VDF-TrFE) is much higher than that of PZT, there will be an electric breakdown across the PZT phase when a high electric field is applied intending to align the dipoles in the P(VDF-TrFE) phase. This indicates that the established fabri-

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cation methods cannot be used to prepare PZT/P(VDF-TrFE) 1-3 composites with both phases piezoelectrically active. As the piezoelectric coefficients (d_{31}, e_{33}, etc.) of PZT and P(VDF-TrFE) are opposite in sign [11], it is expected that the piezoelectric properties of PZT/P(VDF-TrFE) 1-3 composites with both phases poled in opposite directions would be higher than those with unpoled matrix, and those with both phases poled in the same direction would be lower [12]. However, due to the large difference between the piezoelectric coefficients of PZT and P(VDF-TrFE) (e.g., the e_{33} value of PZT is about 60 times larger [11]), the effect of the poled copolymer matrix is significant only for low PZT volume fraction. Therefore, in this work, we aim at fabricating PZT/P(VDF-TrFE) 1-3 composites with low PZT volume percent and with both phases poled either in the same or in opposite directions, and studying their resonance characteristics and transducer performances.

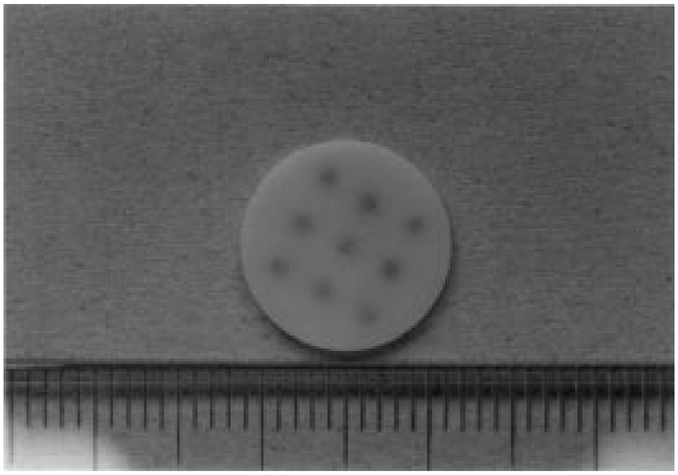

Fig. 1. Photograph of a PZT/P(VDF-TrFE) 1-3 composite with 6 PZT volume percent.

II. Fabrication of 1-3 Composite

Modified PZT powder (PKI-502) of average particle size of 1 μm was used in this work to make the ceramic rods. The powder was supplied by Piezo Kinetics Inc. (Bellefonte, PA) and has properties similar to the Vernitron (Morgan Matroc Ltd., Bedford, OH) PZT-4 composition. The rods were extruded through a die and sintered at 1260°C for 1.5 hours. An extruded unpoled P(VDF-TrFE) (80/20) copolymer sheet supplied by Atochem North America (now Amp Sensors, Valley Forge, PA) was used as the matrix. The sheet was 0.8 mm thick. Its Curie transition temperature for the first heating (Tc↑) and melting temperature were 124.4 and 149.0°C, respectively, indicating a TrFE content of slightly higher than 20% [13]. Before poling, the copolymer sheet was annealed at 120°C for 2 hours to increase the amount of the polar β-phase crystallites of the copolymer. In order to enhance the piezoelectric activities of the copolymer, the sheet was poled using a two-step poling process [14]. The sheet was poled at 105°C in an oil bath for 2 hours, then cooled to room temperature with the electric field kept on; then it was heated again to 105°C and repoled for another 2 hours. The electric field applied to the sheet was 25 MV/m. A poled P(VDF-TrFE) disk of 12.7-mm diameter was cut from the sheet.

An array of holes having the same diameter as the rods were drilled in the poled copolymer sample, in a square pattern with 3 mm center-to-center separation, and the sintered unpoled PZT rods then were inserted into these holes. To glue the rods to the copolymer, the composite was immersed into epoxy (Shell 815 + hardener C in volume ratio 4:1) and degassed in vacuum for 15 minutes. After curing the epoxy for 5 hours at 70°C, the composite was lapped to obtain parallel surfaces. The composite then was cut into a disk of 12.7-mm diameter. Following the same procedure with different rod diameters (0.8 or 1.0 mm), composite disks with 3.6 and 5.6 volume percents of PZT were prepared (Fig. 1). As the piezoelectric coefficients of PZT are much larger than those of the copolymer, we have not prepared composites with larger PZT volume percents, in which the effect of the poled copolymer matrix will be hardly observable, using either thicker PZT rods or smaller rod separation. For comparison, a composite disk with an epoxy (Ciba-Geigy GY9708+HY2992) matrix was also prepared using the same procedure. The epoxy was supplied by Ciba-Geigy, HK in the form of a 3-mm thick plate.

A thin layer of air drying silver paint ($\sim 10\ \mu$m) was applied on each major surface of the composite disk to serve as electrodes. The composite disk was repoled at 110°C under an electric field of 2.5 MV/m in an oil bath for half an hour to align the PZT dipoles. As an electric field of 2.5 MV/m was too low to switch the polarization in the copolymer, the pre-existing piezoelectric properties of the matrix were retained. This was confirmed in the following way: the electromechanical coupling coefficient (k_t) of a poled copolymer disk (prepared following the above procedure) was first measured using the nonlinear regression technique [15]. The poled copolymer disk was then repoled at 110°C under an electric field of 2.5 MV/m for half an hour; and its k_t was remeasured. The k_t of the poled copolymer disk before and after repoling were found to be approximately the same. In order to study the effect of the direction of the polarization of the copolymer matrix, different composite disks were made by poling the PZT rods in a direction either parallel or anti-parallel to the poling direction of the matrix. Poled PZT and P(VDF-TrFE) disks also were prepared. All of the samples used in this work are listed in Table I. Because it is very difficult to pole a thicker P(VDF-TrFE) sample, we have fabricated only PZT/P(VDF-TrFE) composite disks of thickness less than 0.8 mm.

TABLE I
DESCRIPTION OF THE SAMPLES.

Sample	PZT Volume (%)	Rod diameter (mm)	Description
PZT	100	-	poled PZT disk
P(VDF-TrFE)	0	-	poled P(VDF-TrFE) disk
PC4P	3.6	0.8	PZT and P(VDF-TrFE) poled in the same direction
PC4A	3.6	0.8	PZT and P(VDF-TrFE) poled in opposite directions
PC6P	5.6	1	PZT and P(VDF-TrFE) poled in the same direction
PC6A	5.6	1	PZT and P(VDF-TrFE) poled in opposite directions
PE6S	5.6	1	poled PZT, epoxy matrix

III. RESONANCE CHARACTERISTICS OF 1-3 COMPOSITE

Because the wavelength of the shear wave in the matrix, generated by the vibrations of the PZT rods at the longitudinal thickness mode resonance, is not much larger than the rod separation, our composite samples cannot be considered as homogeneous materials [1], [2]. For example, the wavelength of the shear wave at 1.6 MHz (the theoretical longitudinal thickness mode resonance frequency for the PZT rods of 0.80-mm thickness) is about 0.7 mm (shear wave velocity of P(VDF-TrFE) = 1130 m/s, measured using the immersion method for ultrasonic measurement [16]), which is much smaller than the rod separation (3 mm). Therefore, both the rods and the matrix resonate almost independently, and the resonance modes can be classified into two main categories:

1. Resonances of the individual piezoelectric elements:

- The thickness mode (T-mode) resonance of the poled copolymer matrix. Because the diameter-to-thickness ratio of the sample disk is usually large (> 16), the corresponding resonance frequency (f_T) increases linearly with decreasing sample thickness, and can be calculated by assuming no coupling between the vibrations in the thickness and transverse directions.
- The longitudinal thickness mode (H-mode) resonance and the radial mode (L-mode) resonance of the PZT rods. Because the diameter-to-height ratio of the rod is close to 1, these two resonance modes are coupled to each other. The corresponding resonance frequencies (f_H and f_L, respectively) can be predicted from the coupling theory discussed below [7].

2. Cooperative resonances of the composite structure:

- The planar mode resonance (R-mode) of the composite disk. The corresponding resonance frequency (f_R) is determined mainly by the diameter of the composite.
- The stopband resonances (B1- and B2-modes). These resonances result from the interaction between the regularly spaced PZT rods and occur at frequencies near which Bragg reflections occur in the plane of the composite disk. The corresponding frequencies, f_{B1} and f_{B2}, are determined mainly by the periodicity of the PZT rods.

Consider a cylindrical rod resonator of height H and diameter L. The polarization of the sample is aligned in the height direction, and the circular surfaces are entirely electroded. There are two independent vibration modes: the radial mode with frequency f_a and the longitudinal mode with frequency f_b; and the frequencies f_a and f_b are given by [7]:

$$f_a = \frac{\zeta}{\pi L}\sqrt{\frac{c_{11}^E}{\rho}} \tag{1}$$

$$f_b = \frac{X_t}{\pi H}\sqrt{\frac{c_{33}^D}{\rho}} \tag{2}$$

where ρ is the density, c_{ij} are the stiffness constants, the superscripts E and D denote short circuit and open circuit conditions, respectively, ζ is the first positive root of:

$$\Im(\zeta) = 1 - \gamma \tag{3}$$

$$\gamma = 1 - \frac{c_{12}^E}{c_{11}^E} \tag{4}$$

and X_t is the first positive nonzero root of:

$$\tan X_t = X_t / k_t^2 \tag{5}$$

$\Im(\zeta)$ is the modified quotient of Bessel functions [$\Im(\zeta) = \zeta J_0(\zeta)/J_1(\zeta)$] and k_t is the electromechanical coupling coefficient. If there is coupling between the radial and longitudinal modes, the coupled resonance frequencies are given by the solutions of the biquadratic frequency equation [8]:

$$(f_a^2 - f^2)(f_b^2 - f^2) = f_a^2 f_b^2 \Gamma^2 \tag{6}$$

where

$$\Gamma = \sqrt{|(1 - \alpha^2)(\zeta^p/\zeta)^2 - 1|} \tag{7}$$

$$\alpha = \sqrt{(c_{13}^E)^2/(c_{11}^E c_{33}^E)} \tag{8}$$

and ζ^p is the first positive root of

$$\Im(\zeta^p) = \frac{1 - \gamma}{1 - \alpha^2}. \tag{9}$$

The calculated coupled resonance frequencies f at different diameter to height ratios (L/H) will be compared with experimental values in a later section.

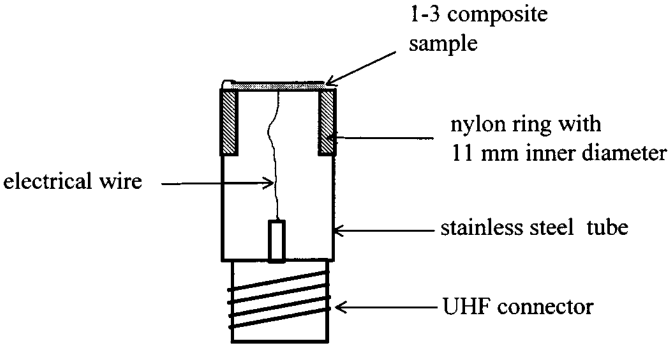

Fig. 2. Schematic diagram of 1-3 composite transducer.

IV. Experiments

A. Resonance Characterization

As all of the resonance modes mentioned here can be excited electrically, an impedance analyzer (HP 4194A) was used to measure the electric impedance of the composite disks as functions of frequency in order to find the resonance frequencies. As the resonance frequencies f_T, f_H, and f_L are dependent on composite thickness, they also were measured as a function of composite thickness, by thinning the composite disks to different thicknesses in steps.

B. Transducer Performance Evaluation

The composite disks were fabricated into transducers with air-backing and with no front face matching layer (Fig. 2), and the pulse-echoes, transmitting and receiving voltage responses of the transducers, were measured.

1. Pulse-Echo Measurements: The experimental setup is shown schematically in Fig. 3. The sample transducer was excited by an electrical impulse of peak voltage −50 V (unloaded) and duration less than 100 ns from an ultrasonic transducer analyzer (Panametrics 5052UA) to generate an ultrasonic pulse in water. The pulse was reflected by a flat stainless steel target at the far field-near field transition region of the wave train [17] and received by the transducer. The echo was acquired by a digitizing oscilloscope (HP 54504A) to obtain a waveform and by a spectrum analyzer (HP 3589A) to obtain an amplitude spectrum. The ringdown, defined as the number of cycles it takes the oscillation to reduce to 10% (−20 dB) of its maximum peak amplitude, was determined from the waveform; and the center frequency f_o and −6 dB bandwidth were determined from the amplitude spectrum. The uncertainty of the measurement arose mainly from the alignment. The uncertainties introduced by the oscilloscope and the spectrum analyzer, after taking an average of 32 measurements, were relatively small ($< 1\%$). However, it was found that the ringdown and f_o were not affected significantly. After repeating the measurement on the same transducer for five times, the reproducibility for the −6 dB bandwidth was about ±2%.

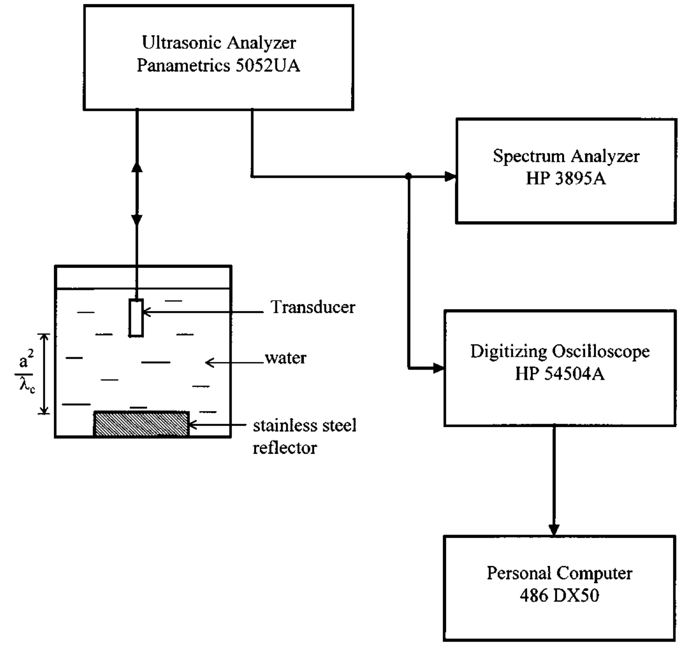

Fig. 3. Schematic diagram of the setup for pulse-echo measurement (spectrum analyzer method). The settings of the ultrasonic analyzer are: Energy = 1, Damping = 50 Ω, Rep. rate = 1 k, Attenuation = 10 dB, Gain = 20 dB, Filter = out.

2. Transmitting Voltage Response Measurements: In the measurement, a function generator (HP 8116A) was used to generate a tone burst of 15 cycles at a single frequency with a repetition rate of 1 kHz (Fig. 4). The signal was amplified by a power amplifier (25A100, Amplifier Research, Souderton, PA) to 15 V (peak), and then was used to drive a sample transducer to generate an ultrasonic wave train in water. The driving voltage (peak) was monitored by a digitizing oscilloscope (HP 54504A). The wave train was received by a PVDF bilaminar shielded membrane hydrophone with an active element of 0.5-mm diameter (Type Y-34-3598, GEC-Marconi Pty., Australia) placed at the far-field near-field transition region of the wave train. The signal from the hydrophone was first amplified five times by a matched amplifier (National Physical Laboratory, Middlesex, UK) and then measured by the oscilloscope. The membrane hydrophone with the 5× amplifier were calibrated by National Physical Laboratory, UK. The acoustic pressure at the hydrophone element was calculated by dividing the measured voltage by the calibrated sensitivities (end-of-cable loaded) of the hydrophone. The transmitting voltage response (P_o) was given by the ratio of the detected pressure to the driving voltage, and expressed in kPa/V. The P_o of a transducer was measured as a function of frequency, from which the maximum P_o was determined.

3. Receiving Voltage Sensitivity Measurements: Following the procedure described here, the acoustic pressure generated by a 10 MHz commercial transducer (0.25-inch diameter, Panametrics V312) at a test distance of 11 cm was first measured as a function of the lateral position by

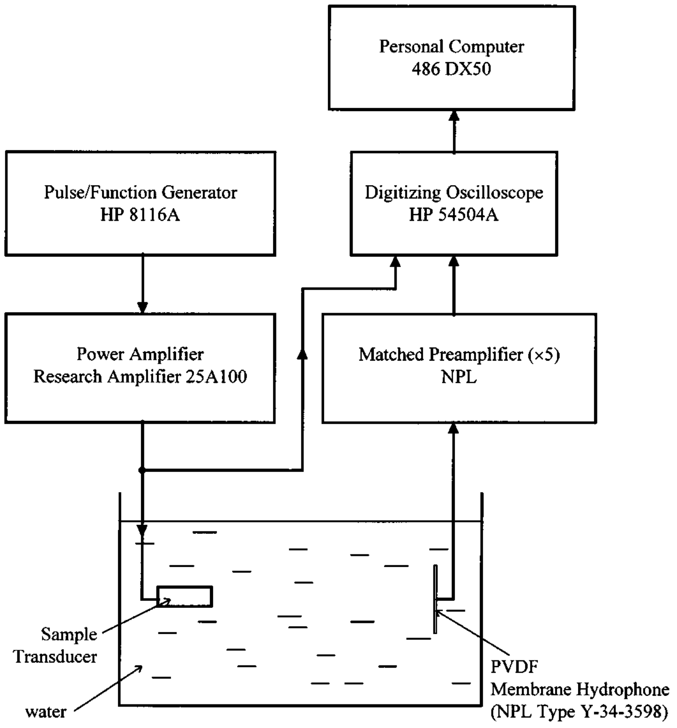

Fig. 4. Schematic diagram of the setup for measuring transmitting voltage response of a transducer.

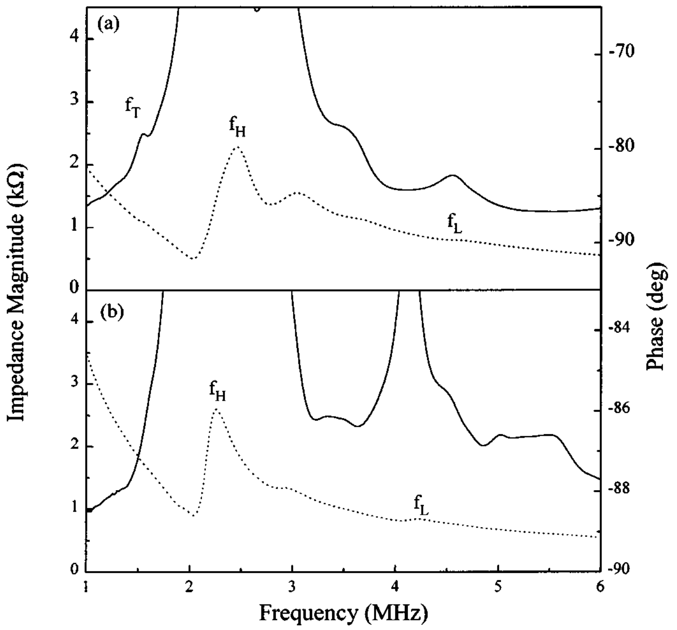

Fig. 5. Frequency plots of the impedance magnitude (- - - -) and phase (——) for (a) PC4P (3.6% PZT, PZT and P(VDF-TrFE) poled in the same direction) of 0.67-mm thickness, and (b) PE6S (5.6% PZT, epoxy matrix) of 0.76-mm thickness.

translating the membrane hydrophone in a direction parallel to the transducer's surface, and an averaged pressure on a surface of 12.7-mm diameter was calculated. The sample transducer then was placed at the same test distance, and the received signal was acquired by an oscilloscope (HP 54504A). The receiving voltage sensitivity (S_o) was calculated by dividing the measured voltage by the averaged acoustic pressure, and expressed in μV/Pa. The S_o of a transducer was measured as a function of frequency, from which the maximum S_o was determined.

In the transmitting and receiving voltage response measurements, the possible errors arose mainly from the uncertainty of the sensitivity of the standard PVDF membrane hydrophone (±7%) and the alignment in obtaining maximum signals. The uncertainties introduced by the oscilloscope and the spectrum analyzer, after taking an average of 32 measurements, were relatively small (< 1%). After repeating the measurements five times, it was found that the reproducibilities for both measurements were about ±5%, so the overall errors in these measurements were about ±7%.

V. Results and Discussion

A. Resonance Characteristics

A number of resonance peaks are observed in the impedance spectra of each 1-3 composite disk at different thicknesses, and the results will be discussed. The resonance peaks associated with the planar mode and stop-band resonances are very weak and far from the other resonance modes, so they are relatively unimportant and will not be discussed.

As compared with the PZT/epoxy composite disk, one more resonance peak attributed to the poled copolymer matrix (T-mode) is observed for the PZT/P(VDF-TrFE) composite disks (Fig. 5). As the k_t of P(VDF-TrFE) is small and its ε_{33}^S is about 150 times lower than that of PZT, the resultant impedance of the two phases connected in parallel in a 1-3 composite is dominated by the PZT phase. As a result, the resonance peak attributed to the copolymer matrix is relatively small and usually overshadowed by other nearby resonances in the impedance spectrum. This is shown in Fig. 6, the impedance spectra of the same composite disk used in Fig. 5 but of smaller thickness, in which the T-mode should be located at 1.90 MHz. Therefore, it is difficult to study the effect of the poled copolymer matrix by means of the impedance spectra.

As the copolymer matrix is assumed to resonate almost independently, f_T (the resonance frequency of the T-mode) should increase with decreasing sample thickness following (2) for the rest of the work. Fig. 7 shows the good agreement between the observed f_T for a P(VDF-TrFE) disk at different thicknesses and the values calculated using (2) and the values of $c_{33}^D = 8.5$ GPa and $k_t = 0.185$ measured using the nonlinear regression technique [15].

The general features of the longitudinal thickness mode (H-mode) resonance and the radial mode (L-mode) resonance at different sample thicknesses are similar for all composite disks with either a P(VDF-TrFE) matrix or an epoxy matrix. Figs. 8(a) and 9(a) show the impedance magnitude and phase as functions of frequency for PC6P

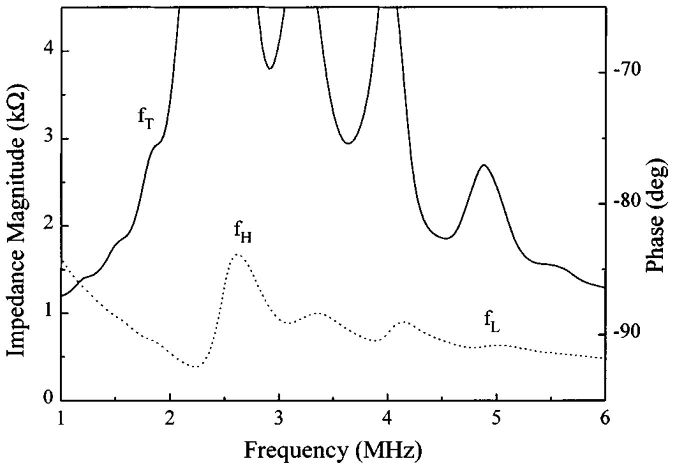

Fig. 6. Frequency plots of the impedance magnitude (- - - -) and phase (——) for PC4P (3.6% PZT, PZT and P(VDF-TrFE) poled in the same direction). The composite disk is 0.56-mm thick and f_T should be 1.90 MHz.

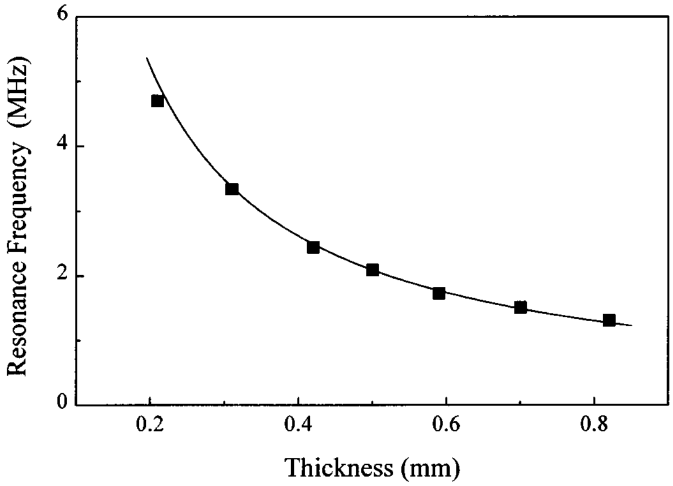

Fig. 7. Variation of f_T with thickness for a P(VDF-TrFE) disk.

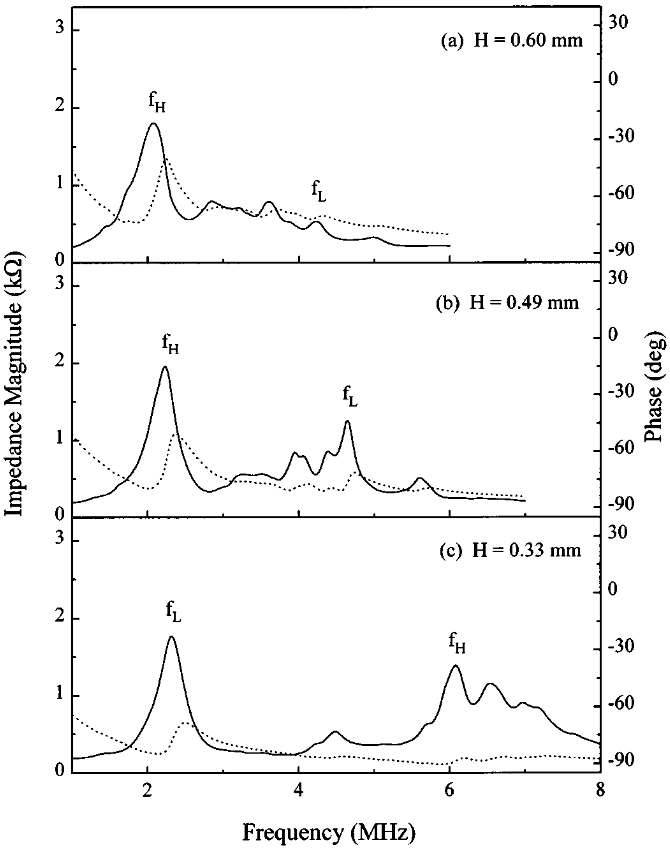

Fig. 8. Frequency plots of the impedance magnitude (- - - -) and phase (——) for PC6P (5.6% PZT, PZT and P(VDF-TrFE) poled in the same direction) at different thicknesses.

TABLE II
Material Parameters of PKI 502.

Parameter	PKI 502
c_{11}^E (GPa)	137
c_{12}^E (GPa)	87
c_{13}^E (GPa)	79
c_{33}^D (GPa)	162
k_t	0.47
ρ (kg/m^3)	7700

and PC6A of thickness 0.60 mm, respectively. With the help of the coupling theory, the strongest mode at 2.0 MHz is identified as the H-mode, and the much weaker mode at 4.3 MHz is the L-mode. There are a few peaks in between. These may be caused by mode coupling of the PZT rods, but the exact reasons are not clear yet. Due to mode coupling, both f_H and f_L increase with decreasing sample thickness, and the two modes cross over, i.e., the H-mode occurs at frequency lower than that of the L-mode, at thickness of 0.40 mm [Figs. 8(a)–(c) and 9(a)–(c)]. The observed f_H and f_L at different sample thicknesses are plotted in Fig. 10 with the values calculated by (6) using the properties of PKI 502 (Table II) measured according to the IEEE Std. 176-1987[1]. There is good agreement between the calculated and observed f_H and f_L, and the direction of the polarization of the copolymer matrix has no significant effect on the resonance frequencies of the rods. This is attributed to the stronger piezoelectricity of PZT, and the relatively softer copolymer that does not impose strong clamping on the ceramic rods.

Fig. 11 shows the impedance and phase spectra for the PZT/epoxy composite disk (PE6S) of different thicknesses, and the calculated and observed f_H and f_L are plotted in Fig. 12. The small discrepancy between the calculated and observed f_H may arise from the clamping imposed on the rods by the hard epoxy matrix. The good agreement between the observed and the calculated resonance frequencies confirms that, although incorporated in a polymer matrix, the PZT rods in the composite vibrate almost independently.

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[1] IEEE Standard on Piezoelectricity, ANSI/IEEE Std. 176-1987.

TABLE III
THE CENTER FREQUENCY f_o, −6 dB BANDWIDTH, RINGDOWN, AND PEAK-TO-PEAK VOLTAGE (V_{P-P}), IN THE PULSE-ECHO RESPONSE FOR THE PZT, P(VDF-TRFE) AND 1-3 COMPOSITE TRANSDUCERS.

Sample	Thickness (mm)	f_o (MHz)	−6 dB BW (%)	−20 dB Ringdown (cycles)	V_{P-P} (mV)
PZT	0.71	3.05	8	21	950
P(VDF-TrFE)	0.54	1.89	33	5	8.6
PE6S	0.60	2.20	16	10	25
PC6P	0.60	1.82	22	9	30
PC6A	0.60	1.93	17	9	54

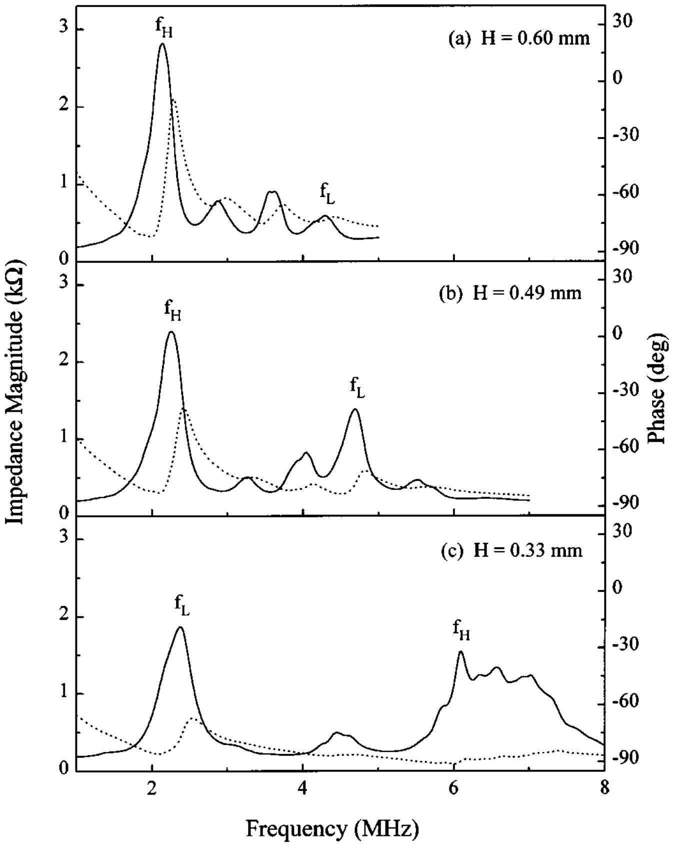

Fig. 9. Frequency plots of the impedance magnitude (- - - -) and phase (——) for PC6A (5.6% PZT, PZT and P(VDF-TrFE) poled in opposite directions) at different thicknesses.

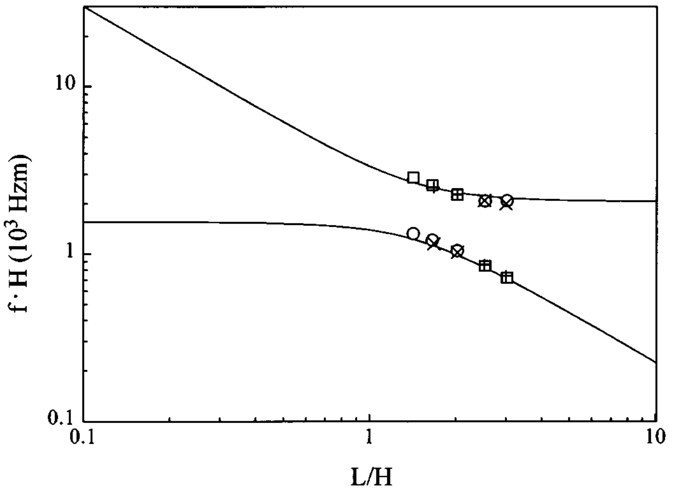

Fig. 10. Log-log plot of resonance frequencies vs. diameter-to-height ratio for PZT/P(VDF-TrFE) 1-3 composites. The solid curves are the theoretical values calculated from (6). Data of PC6P (5.6% PZT, PZT and P(VDF-TrFE) poled in the same direction): f_L, +; f_H, ×. Data of PC6A (5.6% PZT, PZT and P(VDF-TrFE) poled in opposite directions): f_L, □; f_H, ○.

B. Transducer Performance Evaluation

The pulse-echo responses, transmitting and receiving voltage responses of the composites of 0.6-mm thickness, containing 5.6% PZT, were measured. Similar to the observations from the impedance spectra, there is one major resonance peak associated with the H-mode of the PZT rods. At this thickness, for PC6P and PC6A, the peak associated with the T-mode has merged into the main peak. Figs. 13 and 14 show, as examples, the waveforms and amplitude spectra for PC6P and PC6A. The observed resonance frequencies agree with those observed from the impedance spectra. The center frequency f_o, −6 dB bandwidth and ringdown of the echoes for these samples are given in Table III, in which the results for the PZT and P(VDF-TrFE) transducers also are given for comparison. The settings of the ultrasonic analyzer for generating an electric impulse and amplifying the echo (see Fig. 3) are the same for all transducers. Although, because of different impedances, the actual driving voltage for each sample is different, the f_o, −6 dB bandwidth and ringdown of the echoes are not affected significantly. As shown in Table III, the PZT transducer has the narrowest bandwidth and the longest ringdown, indicating that the vibration is only slightly damped. The copolymer transducer has the broadest bandwidth and the shortest ringdown, indicating a heavy damping in this transducer. The performances of the composite transducers are in between these two bounds, and the PZT/P(VDF-TrFE) composite transducers are better than the PZT/epoxy transducer as they have broader bandwidth and shorter ringdown in their pulse-echo responses. This may be attributed to the softness of the copolymer matrix compared to epoxy. In general, for homogeneous resonators, broader bandwidth results in shorter ringdown. However, as the vibrations of

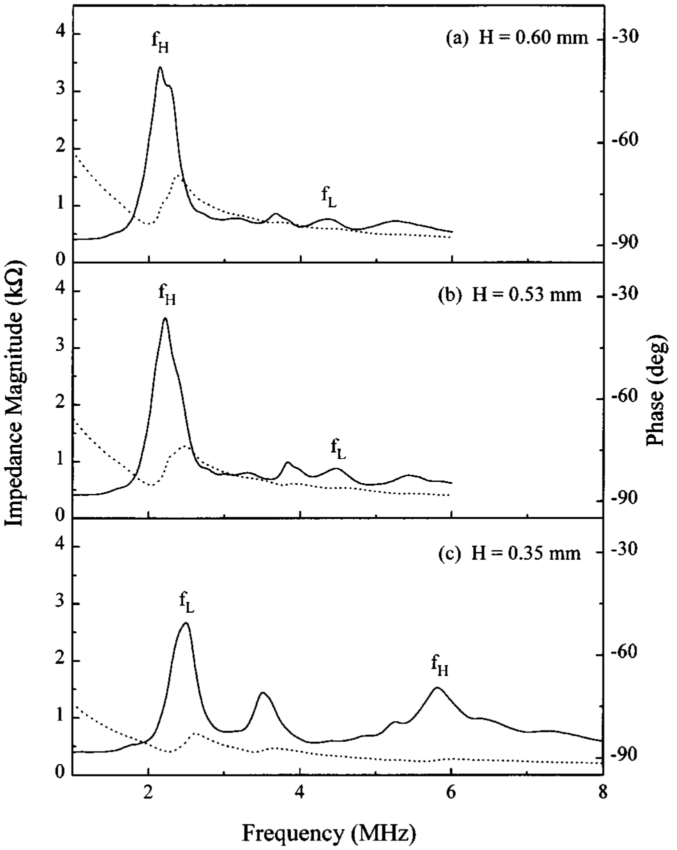

Fig. 11. Frequency plots of the impedance magnitude (- - - -) and phase (——) for PE6S (5.6% PZT, epoxy matrix) at different thicknesses.

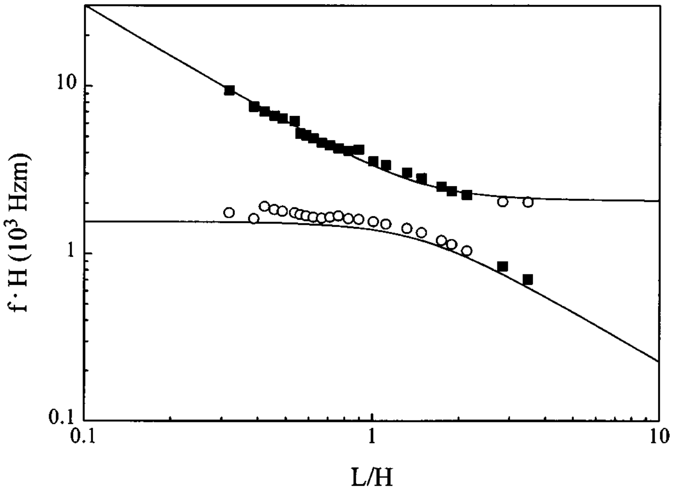

Fig. 12. Log-log plot of resonance frequencies vs. diameter-to-height ratio for PE6S (5.6% PZT, epoxy matrix). The solid curves are the theoretical values calculated from (6). Data: f_L, ■; f_H, ○.

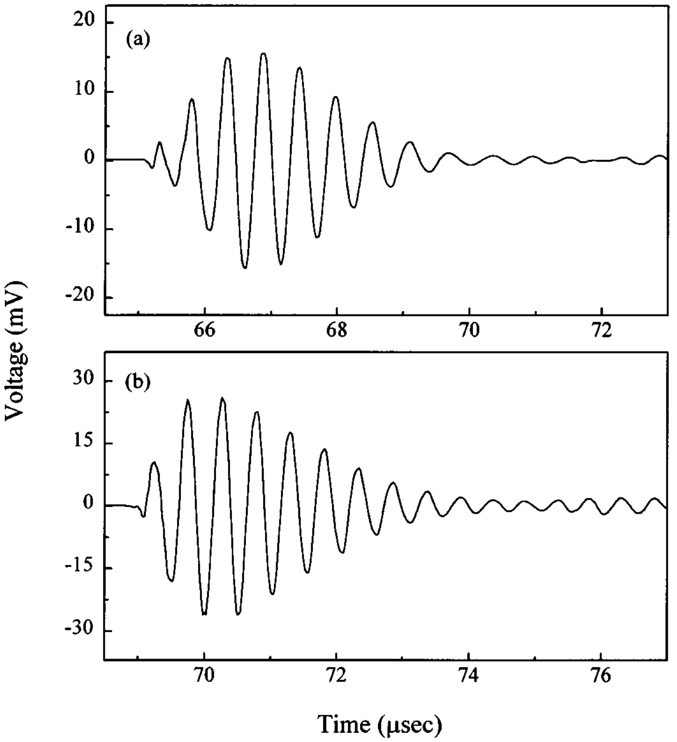

Fig. 13. Waveform of echo for (a) PC6P (5.6% PZT, PZT and P(VDF-TrFE) poled in the same direction) of 0.60-mm thickness, and (b) PC6A (5.6% PZT, PZT and P(VDF-TrFE) poled in opposite directions) of 0.60-mm thickness.

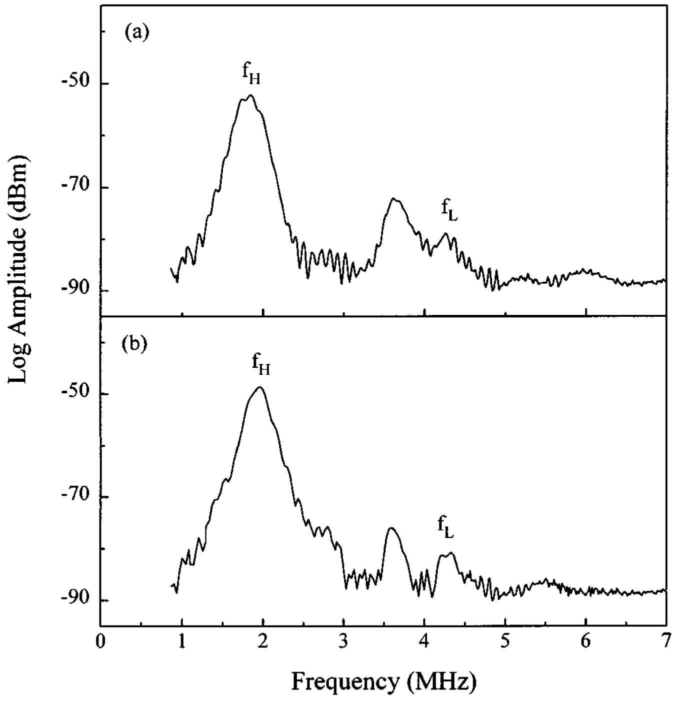

Fig. 14. Amplitude spectrum of echo for (a) PC6P (5.6% PZT, PZT and P(VDF-TrFE) poled in the same direction) of 0.60-mm thickness, and (b) PC6A (5.6% PZT, PZT and P(VDF-TrFE) poled in opposite directions) of 0.60-mm thickness.

the PZT rods are mainly damped down by the copolymer matrix, PC6P and PC6A have the same ringdown, but the bandwidth of PC6P (the two phases are poled in the same direction) is broader than that of PC6A (the two phases are poled in opposite directions). This indicates that the ringdown could not be correlated simply to the bandwidth for composite transducers.

The maximum transmitting voltage response (P_o) and receiving voltage sensitivity (S_o) corresponding to the major resonance peak (H-mode) are listed in Table IV. The maximum P_o and S_o for the PZT and P(VDF-TrFE) transducers are the two bounds, and the PZT/P(VDF-TrFE) composite transducers have higher maximum P_o and S_o, and broader bandwidth than the PZT/epoxy transducer. In order to show the effects of the direction of the polarization of the copolymer matrix, the results for the PZT/P(VDF-TrFE) composite transducers of different composite thicknesses and different PZT volume percents are also listed in Table IV. As shown in Table IV, PC6A of 0.49-mm thickness has the largest values of maximum P_o (4.5 kPa/V) and maximum S_o (28 μV/Pa). For this composite, f_H and f_T are very close ($\sim$ 2.2 MHz, see Fig. 7), that means both the PZT rods and the copolymer matrix resonate at the same frequency. Because the PZT and P(VDF-TrFE) are poled in opposite directions and their piezoelectric coefficients are opposite in sign, their vibrations are in the same direction, and the induced charges have the same polarity. Therefore, in the transmitting mode, the clamping imposed on the PZT rods by the copolymer is the lowest, and the vibration of the rods is the largest, hence giving the largest maximum P_o. In the receiving mode, because both phases are in resonance, the charge induced is the largest that gives the largest maximum S_o.

Comparing all the composite transducers, PC4P of 0.66-mm thickness has low P_o and S_o but has the broadest bandwidths in both the transmitting (49%) and receiving (62%) modes. Although its bandwidth is about 15% narrower, its transmitting voltage response is more than two times higher than that of the P(VDF-TrFE) transducer. For this composite, the PZT volume percent is low (3.6%), so the maximum P_o and S_o attributed to the longitudinal thickness mode of the PZT rods (H-mode) decrease and become comparable to those attributed to the thickness mode of the copolymer matrix (T-mode) (Fig. 15). In addition, the resonance peaks associated with the H- and T-modes just overlap. Hence, the resultant peaks become broader.

When the composites are further thinned down (e.g., thickness = 0.33 mm), the poling direction of the copolymer matrix causes a more significant effect on the resonance characteristics. In a 0.33-mm thick PC6P composite (the two phases are poled in the same direction), three resonance peaks are observed in the frequency plots of P_o and S_o: $f_H \approx 6$ MHz, $f_L \approx 2.1$ MHz, $f_T \approx 3.8$ MHz (Fig. 16). But in a 0.33-mm thick PC6A composite (the two phases are poled in opposite directions), only two resonance peaks are observed (Fig. 17). The peak at about 6 MHz is asso-

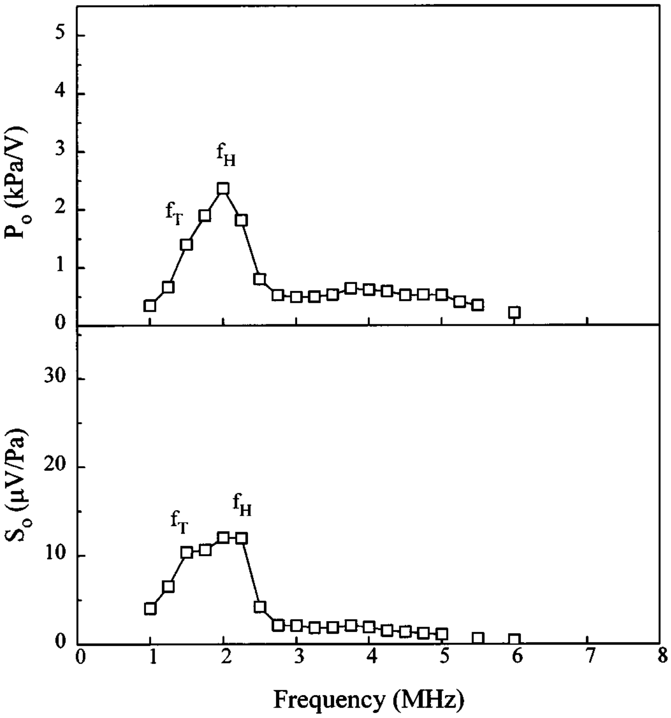

Fig. 15. Frequency plots of (top) transmitting voltage response and (bottom) receiving voltage sensitivity for PC4P (3.6% PZT, PZT and P(VDF-TrFE) poled in the same direction) of 0.66-mm thickness.

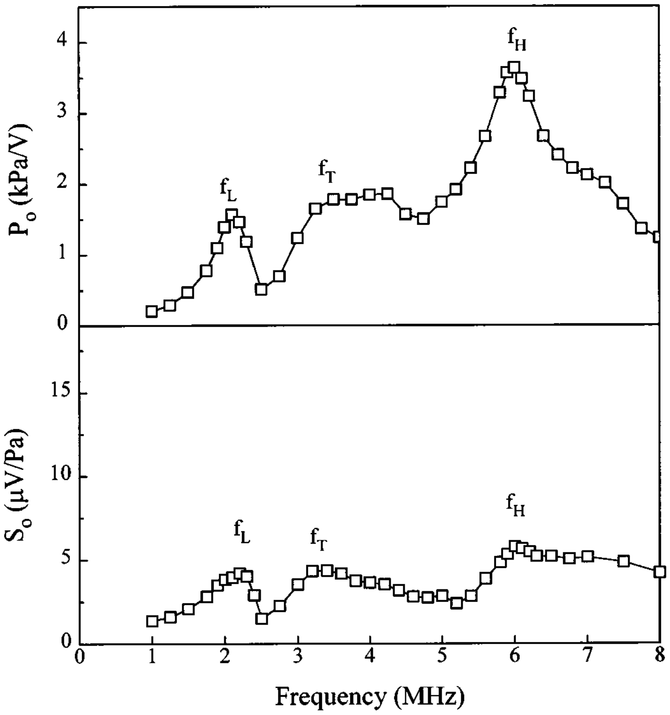

Fig. 16. Frequency plots of (top) transmitting voltage response and (bottom) receiving voltage sensitivity for PC6P (5.6% PZT, PZT and P(VDF-TrFE) poled in the same direction) of 0.33-mm thickness.

TABLE IV
The Transmitting Voltage Response (P_o) and the Receiving Voltage Sensitivity (S_o) for the PZT, P(VDF-TrFE), and 1-3 Composite Transducers.

Sample	PZT Rod diameter (mm)	Thickness (mm)	Transmitting max. P_o (kPa/V)	Transmitting BW (%)	Receiving max. S_o (μV/Pa)	Receiving BW (%)
PZT	-	0.71	71	15	31	14
P(VDF-TrFE)	-	0.54	1.0	54	13	73
PE6S	1	0.60	2.0	29	12	29
PC6P	1	0.60	3.0	32	16	32
PC6A	1	0.60	3.3	32	21	28
PC6P	1	0.49	2.9	27	13	28
PC6A	1	0.49	4.5	33	28	23
PC4P	0.8	0.66	2.4	49	12	62
PC4A	0.8	0.66	2.4	47	18	47

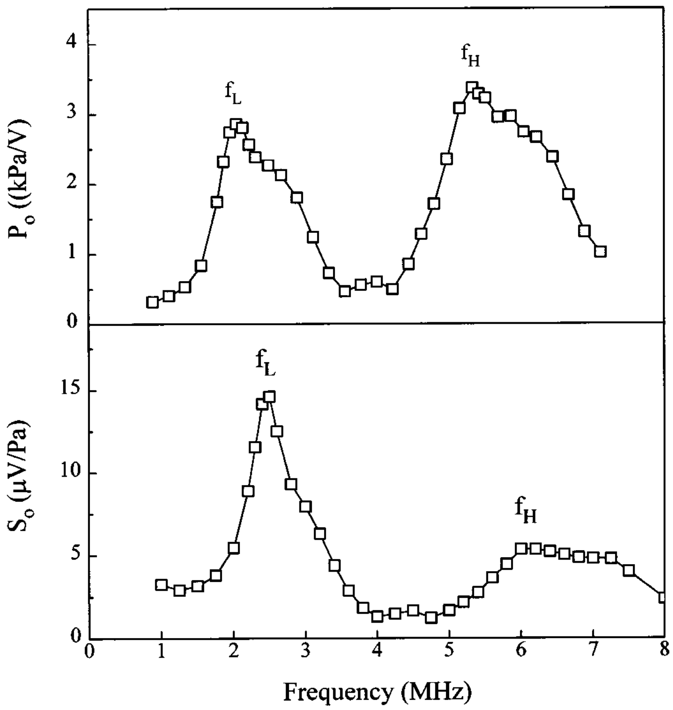

Fig. 17. Frequency plots of (top) transmitting voltage response and (bottom) receiving voltage sensitivity for PC6A (5.6% PZT, PZT and P(VDF-TrFE) poled in opposite directions) of 0.33-mm thickness.

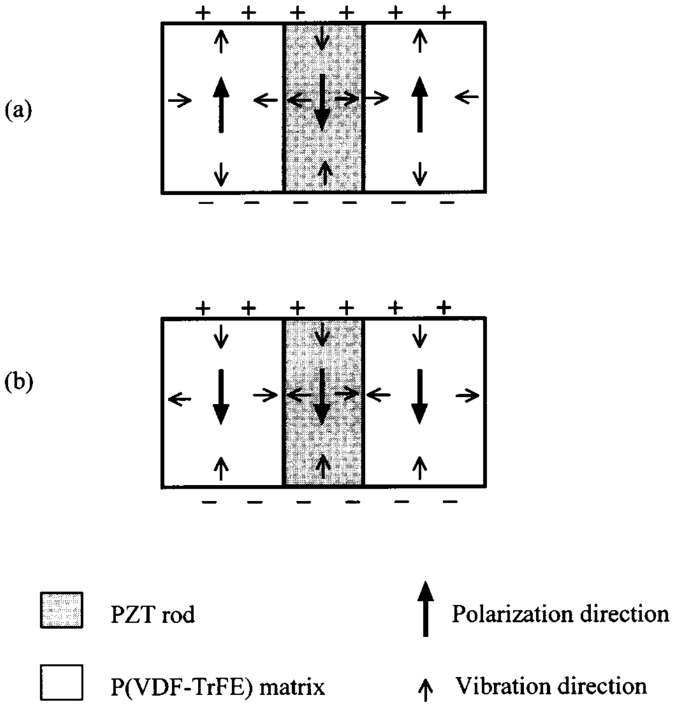

Fig. 18. Vibration directions of the constituent phases of a PZT/P(VDF-TrFE) 1-3 composite near resonance frequency f_L; (a) the phases are poled in opposite directions; (b) the phases are poled in the same direction.

ciated with the H-mode, and the broader peak at about 2.3 MHz results from a merge of the L-mode and T-mode. The merging of the peaks is probably due to the in-phase vibrations of the two phases in the transverse direction. As the exact vibration or displacement pattern of the PZT rods (diameter to height ratio ~ 3) is not known, the postulation is only approximated using the equation for the radial mode vibration of a disk resonator [(11) in [18]]. Accordingly, the PZT rods should vibrate in phase with the driving field in the radial direction at frequencies higher than f_L [Fig. 18(a)]. For PC6A, the vibration of the matrix in the thickness direction is in phase with the driving field, and the vibration in the transverse direction (due to Poisson effect) is 180° out of phase with the driving field [Fig. 18(a)]. So the displacement amplitude or pattern of the two phases in the transverse direction are "cooperative," and the resonance peaks merge together. For PC6P, the vibrations of the two phases in the transverse direction are "against" each other [Fig. 18(b)]. This "against" action is equivalent to stiffening the copolymer matrix, so the T-mode resonance occurs at a higher frequency.

VI. Conclusions

Using a new procedure, PZT/P(VDF-TrFE) 1-3 composites with both phases piezoelectrically active have been fabricated. The dipoles of the two phases are aligned either in the same direction or in opposite directions. Because the separation between the regularly spaced PZT rods is larger than the composite thickness, both the PZT rods and the copolymer matrix vibrate almost independently. The vibration characteristics of the composites is dominated by the longitudinal thickness mode (H-mode) resonance of the PZT rods. The radial mode (L-mode) resonance of the rods is very weak for thick composite disks and becomes significant for thin composite disks (thickness = 0.33 mm). Locations of the observed resonance frequencies of these two modes from the electrical impedance spectra agree well with the values calculated from the coupling theory. Because the resultant electric impedance of a 1-3 composite with two phases connected in parallel is dominated by the PZT phase, the thickness mode (T-mode) resonance of the copolymer matrix hardly can be found in the impedance spectra. Nevertheless, it occurs at a frequency similar to a copolymer disk resonator.

The composite disks have been fabricated into transducers with air-backing and with no front face matching layer, and the pulse-echo response, the transmitting and receiving responses of the composite transducers have been evaluated in water. Performance characteristics of the PZT/P(VDF-TrFE) composite transducers are better than those of the PZT/epoxy transducer. They have a broader bandwidth, and better transmitting and receiving sensitivities. The number of resonances and their frequencies can be adjusted by varying the PZT rod diameter, rod separation, and composite thickness. To fabricate a single-frequency transducer with high transmitting voltage response and receiving voltage sensitivity, it is preferable to pole the PZT rods and the copolymer matrix in opposite directions and have the resonance frequencies f_T and f_H as close as possible, e.g., in a 0.49-mm thick PC6A composite. However, if a single-frequency transducer with broad bandwidth is preferred, the phases should be poled in the same direction, and the resonance frequencies f_T and f_H should just overlap. However, if a multifrequency transducer is desired, the composite disk should be thin enough to have the resonance frequencies f_T, f_H, and f_L well separated, and the ceramic and copolymer matrix should be poled in the same directions, e.g., in a 0.33-mm thick PC6P composite. However, if the phases are poled in opposite directions, a transducer with two operating frequencies at equal efficiencies is obtained, e.g., in a 0.33-mm thick PC6A composite. In conclusion, the PZT/P(VDF-TrFE) 1-3 composites have been shown to be useful ultrasonic transducer materials as it is possible to design 1-3 composite transducers with a variety of performance characteristics. Transducers with multiple operating frequencies should be particularly useful in medical diagnosis, as in many clinical examinations, the patient is examined at several frequencies.

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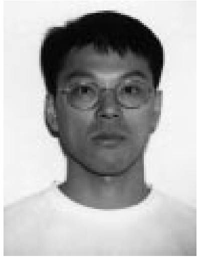

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