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Composite Piezoelectric Materials for Transduction

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ABSTRACT

In order to optimize the detection of acoustical signals in a hydrostatic mode for future applications, new transduction materials which combine active ferroelectric materials with other inert phases such as polymers were developed. These new composite piezoelectric materials exhibit hydrostatic piezoelectric coefficients that far exceed what the conventional lead-zirconate titanate (PZT) ceramics can offer. The particular concept of 0-3 composites is discussed in detail as a typical example of recent accomplishment in this area of new materials technology. The dielectric and piezoelectric properties of some 0-3 composite samples are given as a function of temperature and pressure. The acoustic performance of a prototype hydrophone array is also presented to demonstrate the potential of these materials for future transducer applications.

INTRODUCTION

The development of hull-mounted conformal arrays for acoustical detection has been emphasized for some time. New submarines with higher speeds and greater depth capability require hydrophones in their sonar arrays to perform in the presence of flow-induced acoustic noise. This noise is mainly represented by the convective wavenumber peak of the energy spectrum of a turbulent boundary layer.[1] The frequencies related to the convective components may be relatively high, compared with the acoustical frequencies of interest. But, to a broadband hydrophone the energy contained in those convective components could easily be a major source of self-noise of the array. This self-noise, of course, is expected to interfere destructively with the acoustical signal to be detected and

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therefore reduce the signal-to-noise ratio of the array. A potential solution to this array noise problem is to use large-area hydrophones and two-dimensional beam forming for broadband tracking while keeping the effects of flow-induced noise to a minimum. Large-area hydrophones offer the advantage of allowing incoherent cancellation of the convective-peak-dominated flow noise. Since this noise has only short correlation lengths associated with high wavenumbers, the flow noise effect can thus be minimized.

New piezoelectric materials in the form of large, flexible sheets are ideally suited for the fabrication of large-area hydrophones. These materials need to have high sensitivities in a hydrostatic mode, since all surfaces of the hydrophone element in a hull-mounted conformal array will be exposed to the acoustical pressure. Furthermore, the hydrophone material should be light in weight, as the array may cover an extensive portion of the surface of the submarine. Piezoelectric PVDF thick-film polymers were considered earlier[2,3] for this application, but were found to be limited by the temperature dependence of their piezoelectric properties and by their inherently low dielectric constants. New piezoelectric composites that exhibit high hydrostatic-mode sensitivities are therefore sought as alternatives to PVDF.

Hydrostatic mode sensing

The sensitivity of a hydrophone material in a hydrostatic mode is best described by the piezoelectric voltage coefficient g_h:

$$g_h = g_{33} + g_{32} + g_{31} \tag{1}$$

where the subscript 3 denotes the poled direction of a sample with a pair of electrodes on two surfaces across its thickness. The coefficient g_{33} is the thickness-mode response, and g_{32} and g_{31} are the responses in the plane of the sample. For conventional PZT ceramics, the g_{32} and g_{31} coefficients are negative, and their magnitudes are just about equal to one half that of the g_{33} coefficient.[4] The net result is that the g_h coefficients for PZTs are small, (see Table 1). To design new materials that have high g_h coefficients, clearly one has to minimize g_{31} and g_{32} and/or increase the g_{33} coefficients.

The concept of piezocomposites

The basic concept for the design of piezoelectric composite materials is to develop multiphase materials by combining the desirable properties of several component phases in a composite through the selection of proper

TABLE 1
Piezoelectric Properties of Some Transduction Materials

Material	$\rho (10^3\ kg/m^3)$	K_{33}^T	$\tan\delta (\%)$	$d_{33}\ (pC/N)$	$d_h\ (pC/N)$	$g_h\ (mV\ m/N)$
PZT-4	7·6	1 300	0·5	289	43	3
PZT-5	7·6	1 700	2·0	374	15	2
PVDF	1·4	11	1·3	34	14	152
0–3 Composites						
PR-303	5·3	43	6	48	17	45
PR-304	5·3	40	3	56	19	55
PR-305	5·5	37	3	46	41	124
PR-306	5·3	38	2	34	20	58
PR-307	5·9	45	5	52	44	111

configurational patterns for each phase. Improved properties can be achieved by using, say, a ferroelectric ceramic phase and a polymeric phase. Newnham *et al.*[5] at the Materials Research Laboratory of the Pennsylvania State University pioneered this concept under the sponsorship of the Office of Naval Research. Material properties are controlled by the manipulation of a design parameter called the 'connectivity', which describes the manner each component phase connects itself in the composite in either zero, one, two or three dimensions. The introduction of an inert polymeric phase essentially decouples the lateral Poisson stresses in the bulk ceramic, resulting in a reduction of the contributions of the 31 and 32 terms in Eqn (1). Properties of new composites with connectivities of 3-1, 3-2, 3-3, 1-3 and 0-3 were investigated previously[6,7]. Figure 1 gives a most vivid illustration of the advantage of a composite design in providing superior hydrostatic-mode performance. In this case, the 3-1 composite is represented by a PZT block with holes drilled through in one direction perpendicular to the poled direction of the PZT. The holes were subsequently back-filled with a selected polymer resin such as a room-temperature cured liquid epoxy. The PZT has a connectivity of '3' and the polymer phase has a connectivity of '1', hence the term 3-1 composite. It is seen from Fig. 1 that the hydrostatic d_h coefficient was determined to be about 220 pC/N, as compared with 20 for a homogeneous PZT-5 sample. The hydrostatic g_h coefficient was also dramatically increased from 3 mV-m/N for PZT-5 to about 70 mV-m/N. Over the frequency range of 50–1000 Hz the properties were nearly constant. For hydrostatic pressures up to 7 MPa the material was also stable. Therefore, by simply introducing a one-dimensional polymeric phase in the transverse direction, one was able to reduce the 31 and the 32 components in Eqn (1) that adversely affect the hydrostatic-mode

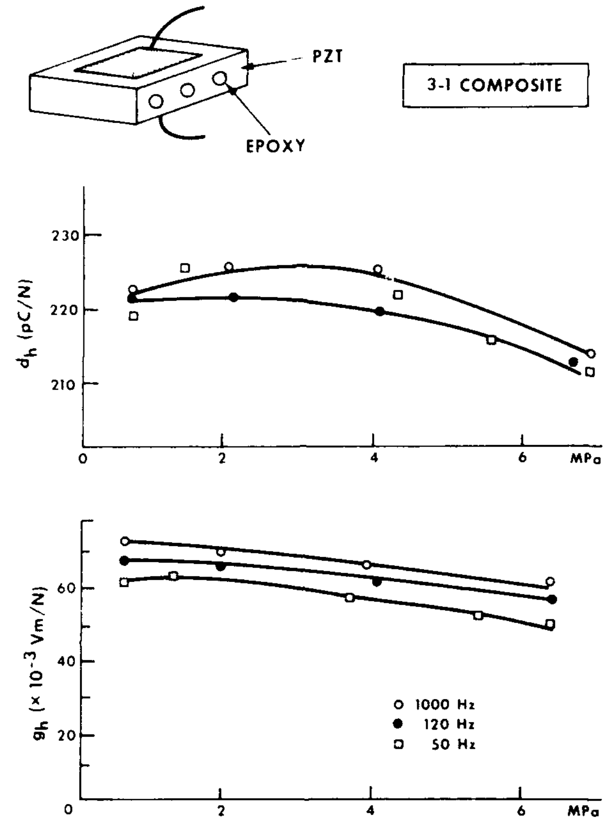

Fig. 1. Piezoelectric d_h and g_h properties of a 3-1 composite sample as a function of pressure.

response, resulting in an improvement in the d_h and g_h properties by more than an order of magnitude.

0-3 Piezocomposites

The 0-3 composites generally contain high volume fractions of fine ceramic particles uniformly dispersed in an elastomeric matrix. The nomenclature of '0-3' is derived from the '0' connectivity for the individually isolated particles and the connectivity of '3' for the elastomer phase that is connected in all three dimensions. The NTK Technical Ceramics Division of the NGK Spark Plugs Corporation in Japan manufactures a series of 0-3 composites under the trade name Piezo-Rubber.[8] Similar types of materials were also investigated by Plessey Research Ltd in the UK.[9] The

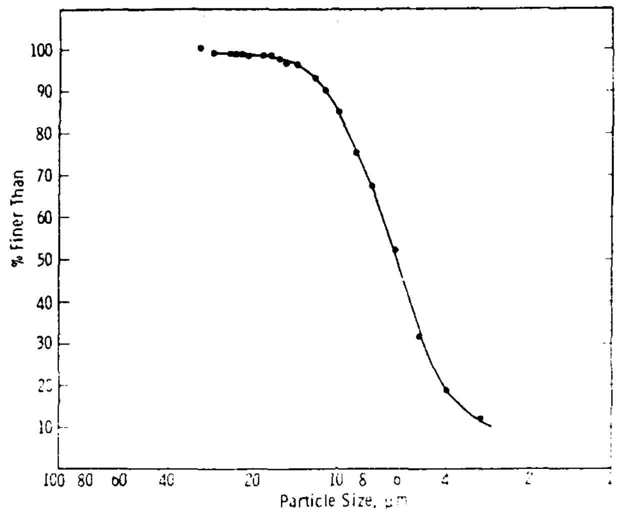

Fig. 2. Particle distribution in a 0-3 piezocomposite sample.

NTK composites studied here contain carefully designed lead titanate particles in a neoprene rubber matrix. The ceramic particles have a very narrow particle size distribution, as shown in Fig. 2. Good bonding between the ceramic particle and the rubber is important for achieving successful poling and good piezoelectric properties. Figure 3 shows the g_h coefficients of an NTK 0-3 composite sample measured at 1 kHz as a

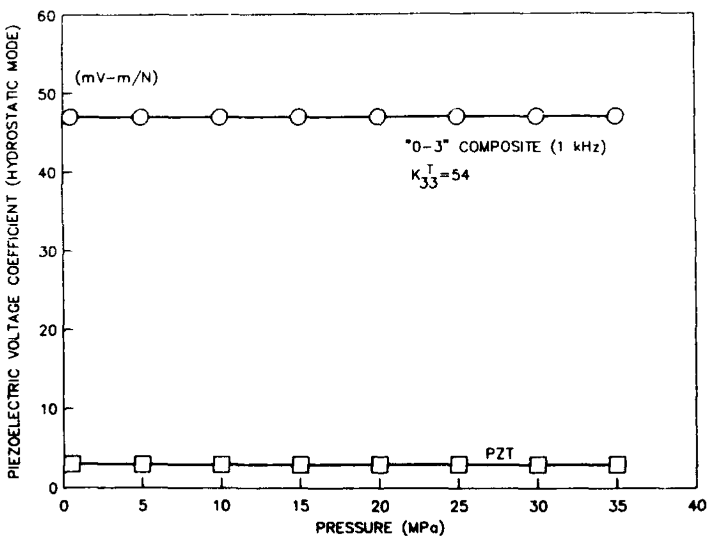

Fig. 3. Piezoelectric g_h coefficient of a 0-3 piezocomposite sample as a function of pressure.

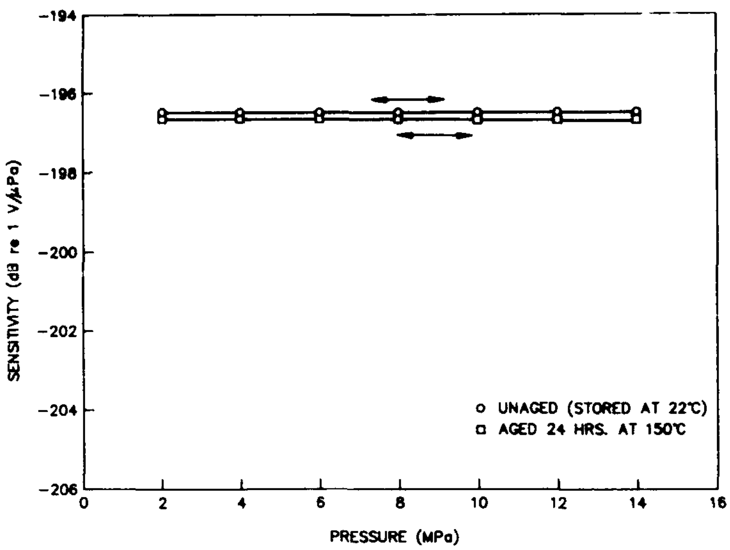

Fig. 4. Aging properties of a 0-3 piezocomposite sample for 24-hour aging at 150°C.

function of hydrostatic pressure. The g_h value of about 50 mV-m/N was almost 20 times that for a typical PZT ceramic, and was stable up to a pressure of 35 MPa. Since the hydrostatic sensitivity of a hydrophone is equal to the product of g_h and the thickness of the sensing element, the pressure-stable g_h property of this 0-3 composite suggests that it is a promising candidate for the design of planar hydrophones. The properties of several types of Piezo-Rubbers[10] representing different amounts of ceramic loading are given in Table 1 along with the properties of other piezoelectric materials for comparison. It may be noted that the g_h coefficient for PR307 Piezo-Rubber is comparable to that of PVDF polymers, but Piezo-Rubber can be easily fabricated to several millimeters in thickness, a limitation that PVDF technology has not been able to overcome. The superior hydrostatic sensitivity of the 0-3 composites together with their low density in comparison with solid PZTs give them distinct advantages over the conventional ceramics for planar hydrophone applications. Their pressure stability, rubber-like formability, and the possibility of controlling their dielectric properties represent additional advantages. Recent progress in the modeling and property modification for 0-3 piezocomposites has been reviewed by Banno.[11]

Figure 4 shows the hydrostatic-mode acoustic sensitivity of a 0-3 composite sample measured at 1 kHz over the pressure range of 2–14 MPa. The sample thickness was 3 mm. It may be noted that the good properties of this composite material was maintained after the sample was aged in an air-circulated oven at 150 C for 24 h. This aging stability of the rubber–ceramic composite is usually not found in conventional PVDF polymers.[12]

HYDROPHONE DEVELOPMENT

Piezo–Rubber tiles with a nominal dimension of 6.35 cm × 6.35 cm × 3 mm were cut for the fabrication of hydrophones. The material was supplied with a conductive layer bonded to the sample as the electrode. Two tiles were bonded together, one on top of the other, and electrically connected in parallel to form one single hydrophone element. This bilaminar design has the advantage of cancelling out any planar responses in the hydrophone due to unwanted structural vibrations, so that the element will function purely as a sensitive hydrostatic device. A 1 × 4 array was assembled and encapsulated by using Conap EN-12 polyurethane compound for water-proofing. The array was acoustically calibrated in the Naval Research Laboratory's anechoic tank facility (ATF). Free-field voltage sensitivity was measured as a function of frequency and pressure. Directivity patterns and the relative phase between adjacent elements were also determined.

ACOUSTIC PERFORMANCE

Figure 5 shows the free-field voltage sensitivity of the 0-3 piezocomposite hydrophone array obtained in the ATF. The constant response over the frequency range of 2–50 kHz was independent of pressure up to 7 MPa. This response is ideal for application as a broadband receiving array. The lower end of this testing frequency range at 2 kHz was bounded by the limit of the size of the anechoic tank. The sensitivity is expected to remain constant at lower frequencies, where the acoustic wavelengths are much longer than that the device dimensions.

Figure 6 shows the directivity patterns of hydrophone elements at 5, 10, 20, 50, and 100 kHz. The positions of the side lobes and the depth of the null are all in excellent agreement with theoretical predictions[13]. This is a good indication that the piezocomposite hydrophones behave acoustically as expected. Figure 7 shows the hydrophone response to an oncoming acoustic wave illuminated from the 'end-fire' position. Theoretically, the response should obey[13] a sinc function ($\sin x/x$) as indicated by the dotted curve in the figure. The experimental result again was very satisfactory.

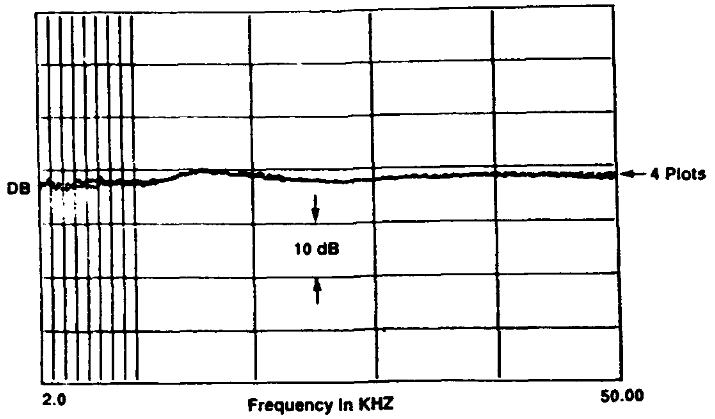

Fig. 5. Free-field voltage sensitivity of −196 dB re V/μPa measured for 0-3 piezocomposite hydrophones from ambient pressure up to 7 MPa.

Furthermore, it is important to ensure that the inter-element coupling between hydrophones in the array be as small as possible. In other words, each hydrophone should behave as a point element so that proper beam-forming can be achieved. Specifically, upon acoustical illumination, one would desire that the relative phase difference between two elements follows the ideal phase difference for two uncoupled point elements located at the centers of the large-area hydrophones. As a measure of the inter-element coupling in the array, the phase of one hydrophone relative to its adjacent neighbor was therefore determined as a function of

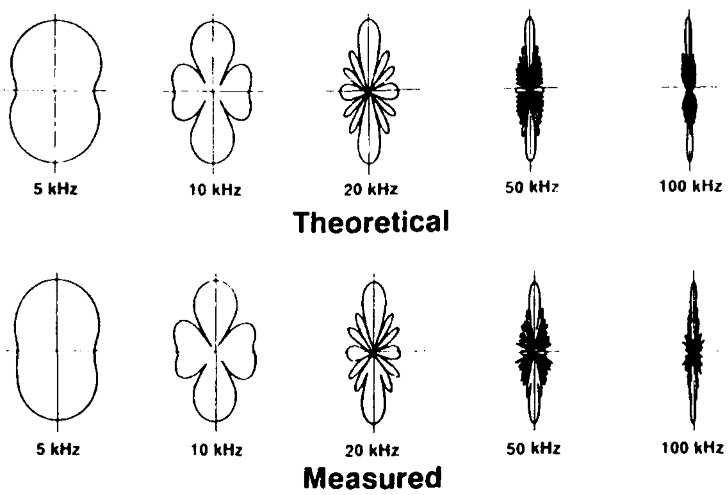

Fig. 6. Directivity patterns of 0-3 piezocomposite hydrophones.

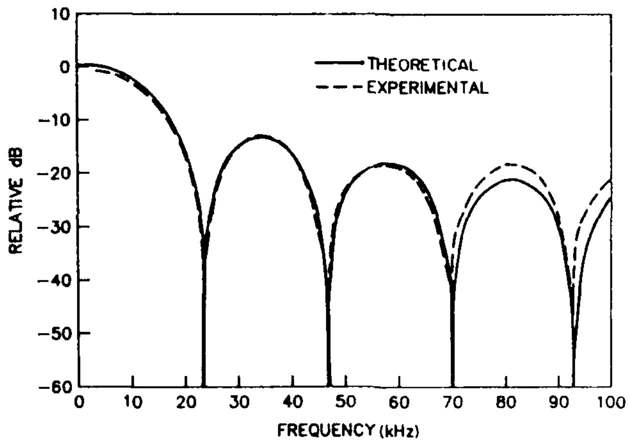

Fig. 7. The end-fire response of 0-3 piezocomposite hydrophones, following the theoretical sinc function.

frequency. If the array is positioned at an angle θ relative to a sinusoidal sound source used to ensonify the array in the experiment, and the hydrophones are separated by a center-to-center distance d, then the ideal phase difference in such an arrangement for two uncoupled point receivers is[13]

$$\text{Phase } (°) = (f\, d \sin\, \theta/c) \times 360 \qquad (2)$$

where c is the sound speed in water and f the frequency. For a given d and θ, Eqn (2) shows that the relative phase is linearly proportional to the frequency.

In Fig. 8, the relationship of Eqn (2) is given as the solid line, together with the experimental data obtained for the 0-3 composite hydrophones. The result, presented here for the case of $\theta = 45°$, is in excellent agreement with the theory over the frequency range tested. In comparison, when two PVDF tiles were evaluated in a similar fashion, the relative phase difference deviated from the theoretical prediction by as much as 40°.[3] This deviation may be attributed to the high values of the piezoelectric 31 coefficient in PVDF. The PVDF polymer is processed by extrusion followed with a uniaxial stretching and poling. The resulting sample will exhibit a very strong planar anisotropy; namely, a very large g_{31} but a small g_{32} coefficient. In contrast, the piezoelectric g_{31} and g_{32} coefficients for the 0-3 composite samples are small in value, and are also equal to each other. It is believed that the good performance of low inter-element coupling in the piezocomposite array is related to this property.

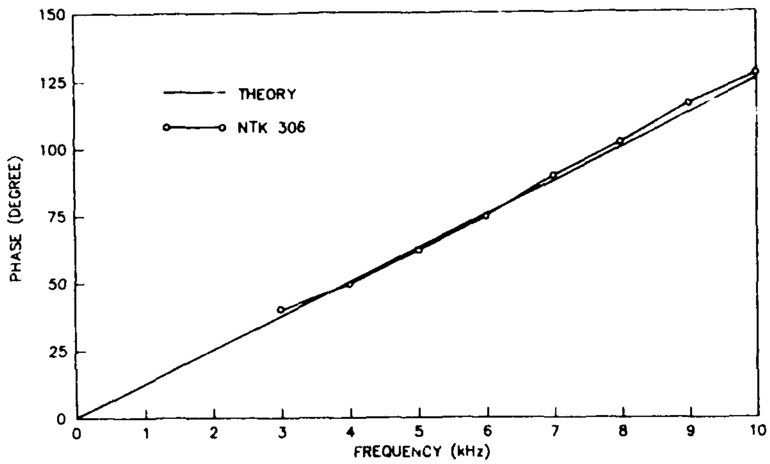

Fig. 8. Relative phase between two 0-3 piezocomposite hydrophones as a function of frequency obeys the theory of Ref. 13.

CONCLUDING REMARKS

Piezoelectric properties of new 0-3 composite materials are presented as an example to show the potential of property optimization through composite designs for hydrophone application. These composite materials exhibit high piezoelectric g_h coefficients, moderate dielectric constants, and low planar piezoelectric coefficients. In comparison, PVDF polymers exhibit a large piezoelectric g_{31} coefficient. The low dielectric constant of PVDF also results in a very low capacitance for most devices, which in turn requires expensive built-in electronic packaging and elaborate water protection for use in hydrophones. The higher dielectric constant of the 0-3 Piezo–Rubber composites greatly relieves these limitations. Since the Piezo–Rubber type 0-3 composites are elastomer-based materials, it is very easy to handle during the fabrication process, and can be conveniently water-proofed by using commercially available polyurethane resins. On the contrary, bonding was very difficult in the fabrication of PVDF hydrophones, because the polymer is a highly fluorinated compound. PVDF does offer the advantages of being lighter weight, mechanically strong and flexible, and having a good impedance match to water. Piezo–Rubber samples have a specific gravity of more than five, greater than ideal for hull-mounted array applications. In its present form, this composite also shows another shortcoming of having a high dielectric loss factor for some applications (about 6%). However, this may be correctable by modifying the elastomer formulation used in the design of the 0-3 piezocomposites.

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