



## Solid Freeform Encapsulation of 1-3-0 PZT Composites

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**Abstract.** Two types of encapsulated lead zirconate titanate (PZT) composite hydrophones have been built and tested: a straight walled 1-3-0 PZT composite design and a curved wall 1-3-0 PZT design. The solid freeform fabrication (SFF) technique was used to construct the polymer encapsulation for the two prototype designs.

In the 1-3 composites ‘soft’ PZT bars are embedded in a polymer matrix with a surrounding air gap. The polymer encapsulations were fabricated on a SFF stereolithography machine in order to enclose the gap and the PZT. Air gaps between the polymer and the PZT were shown to increase the hydrostatic piezoelectric response ( $d_h$ ) by absorbing the lateral hydrostatic stress. Because of the small percentage of PZT, these composites had a low density (neutral buoyancy), low dielectric constant, and, therefore, a lower dielectric constant, and a corresponding higher hydrostatic voltage coefficient,  $g_h$ .

The curved-wall composite design decoupled the PZT component more effectively from the lateral pressure, and thereby improved upon the direct piezoelectric effect. The large cap surface and the curved-wall further improved the amplification of the mechanical loading on the PZT element. The effective hydrostatic charge coefficient  $d_h$ , reached about 1100 pC/N which amplified the  $d_{33}$  coefficient, and greatly enhanced the  $d_h g_h$  figure of merit.

### Introduction

Many hydrophones use piezoelectric ceramic as the active material for low-frequency acoustic wave transduction [1]. The sensitivity of a hydrophone is determined by the voltage that is produced via hydrostatic pressure applied to the transducer [2]. Hydrostatic piezoelectric voltage coefficient ( $g_h$ ) can be determined from the following equation [3]:

$$g_h = d_h / K_{33} \epsilon_0 \quad (1)$$

where  $d_h$  is the piezoelectric hydrostatic charge coefficient,  $K_{33}$  is the dielectric constant in the poling direction, and  $\epsilon_0$  is the permittivity of free space. The product of the  $d_h$  and  $g_h$  coefficients is used as the figure of merit for evaluating a material for use as a hydrophone. A high  $g_h$  coefficient and low dielectric constant are desirable for a sensitive hydrophone. Furthermore, the piezoelectric element within the device should match the acoustics of the water. The hydrophone should also

be rugged enough to withstand mechanical shock from sudden pressure fluctuations [3].

Among many possible piezoelectric materials, lead zirconate titanate (PZT) has been extensively used in hydrophones but it also exhibits several disadvantages [1, 3]. PZT has a very high value of the piezoelectric charge coefficient, but the hydrostatic coefficient  $d_h (= d_{33} + 2d_{31})$  is very low because  $d_{33}$  and  $d_{31}$  have opposite signs and  $d_{33} \approx -2d_{31}$ . The hydrostatic  $g_h$  coefficient is also low because of the high dielectric constant of PZT-5H [3]. These decrease the value of the figure of merit  $d_h g_h$ . Piezoelectric composites, which consist of a piezoelectric ceramic in an inactive polymer, have been the focus of much research work in the last two decades in order to overcome these disadvantages. Numerous devices and structures with various arrangements and connectivity have been studied and explored for different applications [3]. Among a variety of the approaches, piezoelectric transducers with enclosed void show several special advantages, including low acoustic impedance, reduced mass, sensitivity

to weak hydrostatic waves, and enhanced displacement through flextensional and rotational motion [4]. Some piezocomposites with enclosed void show exceptional hydrostatic sensitivity [5–8].

In order to assist in the fabrication of these devices, computer aided design (CAD) and manufacturing (CAM) techniques have been enlisted. Solid Freeform Fabrication is the machine capability to convert computer aided designs (virtual objects) to solid objects without part-specific tooling. It offers the opportunity to build near net parts with a variety of materials and different designs. The application of this technology also appears to have potential in possible small scale production of parts having complex and unique geometries [9].

This paper focuses on techniques used in the fabrication of the polymer encapsulation for piezoelectric ceramic composites via the SFF technique. SFF was applied solely to the construction of the polymer walls surrounding the PZT in order to achieve an improvement in the performance of hydrophone. Hydrostatic piezoelectric properties of these structures have been evaluated.

### PZT—Polymer Composite

A 1-3-0 type piezoelectric ceramic–polymer composite was built as a  $4 \times 4$  matrix with 16 embedded PZT bars acting as active elements. The polymer matrix serves as the supporting structure with air gaps around the PZT bars partially isolating them from lateral stresses. The polymer matrix also adds flexibility to the structure and reduces density and dielectric constant. A schematic of the 1-3-0 composite is shown in Fig. 1.

A CAD program was used to design the polymer matrix, which was then built on a stereolithography machine with a CAD generated. STL file. This unit was supplied by 3D Systems of Valencia, CA. A proprietary formulated SL 5170 photo-curable polymer was used in these builds. Each layer was green cured via a ‘low’ energy laser exposure and then each subsequent layer

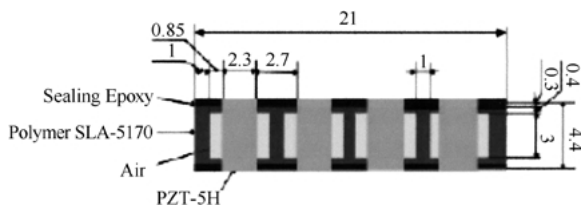


Fig. 1. Schematic of the 1-3-0 composite. units: mm.

was applied above the previous layer. The final geometry was, thereby, ‘built-up’ from two dimensional planar slices. After the initial exposure, the laser-generated matrix was cured for another 24 hours.

Each 15-gram mixture was pressed into a  $25 \text{ mm} \times 25 \text{ mm} \times 5 \text{ mm}$  rectangular pellet under a pressure of  $\sim 56 \text{ MPa}$  perpendicular to a large flat surface. During sintering, the plates were heated from room temperature to  $1300^\circ\text{C}$  for 1 hour [10]. In order to make up for the volatilization of the PbO from the PZT, 5–8 grams of lead zirconate powder was spread across the bottom of the crucible before sintering [10]. To avoid the problems of structural defects in the PZT-5H bulk, the sintered PZT-5H was cut in half lengthwise and poled separately. The PZT-5H pellets were then cut again with a diamond saw into smaller bars and polished to obtain flat surfaces. After cutting and polishing, the PZT bars were about 3–4 mm longer than the thickness of the polymer matrix. The poled PZT bars were then inserted in the polymer matrix with 1.5–2 mm extending on each side of the matrix. A thin layer of fast drying epoxy (Devcon, Desplaines, IL) was applied to the two large surfaces of the composite and the PZT bars were displaced back and forth slightly to permit the epoxy to fill the interface between the PZT and polymer matrix, thereby ensuring the quality of the seal. The encapsulated parts were put in air for 5 minutes to let the epoxy dry. A vat of two-part epoxy (METLAB Epoxy Kit, Metlab, Niagara Falls, NY) was prepared and the dried parts were placed into the vat in order to cover the top and the bottom of the composite. To retain the air gaps in the structure (and to remove bubbles from the epoxy), the vat was placed into a vacuum. Afterwards, the vat of epoxy, with parts inside, was cured in air for 24 hours increasing the reliability of the seal between the air gaps and the surface. After the curing, all composite parts were removed, cut and polished into the final shape.

An electrode material (Ablestik FSCM 21109, Electronic Materials & Adhesives, Rancho Dominguez, CA) was applied on the two main surfaces of the composites, then put into an oven at  $70^\circ\text{C}$  for 1 hour to cure the epoxy. Two wires were attached onto each side of the electrode area using the same electrode material. The assembled 1-3-0 composite is illustrated in Fig. 2.

The  $d_{33}$  coefficient was measured at a frequency of 100 Hz using a Berlincourt  $d_{33}$  meter. Depending on the sample size and the arrangement of PZT bars, 64 measurements were made on each sample. Typically, a grid was drawn on the surface and measurements

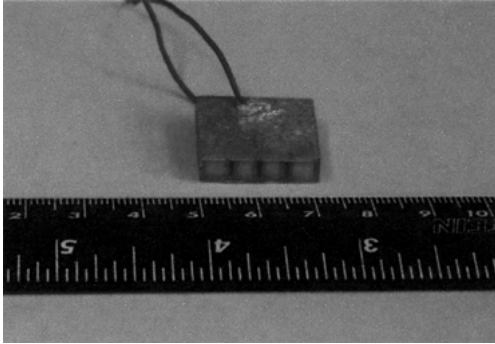


Fig. 2. Fully assembled 1-3-0 composite.

were taken at the center of each of the grid squares. The average of the 64 readings was regarded as the  $d_{33}$  coefficient. The dielectric permittivity was determined from the capacitance at 400 Hz using an impedance analyzer. Average values of  $d_{33}$  and relative dielectric constant from nine samples were 352 pC/N and 540, respectively. The difference between the experimental and theoretical values of  $d_{33}$  for PZT-5H may result from insufficient stress transfer between the polymer and the PZT rod. This effect could be minimized by reducing the distance between the PZT bars or by using a face plate.

A hydrostatic pressure chamber was used to measure the hydrostatic piezoelectric coefficient,  $d_h$ . The hydrostatic piezoelectric properties were plotted as a function of hydrostatic pressure in Fig. 3(A) and (B).

As shown in Fig. 3(A), at pressures above  $\sim 0.7$  MPa (100 psi), the average  $d_h$  and  $g_h$  values of the hour samples stabilized at about 145 pC/N and  $32.7 \times 10^{-3}$  Vm/N, respectively, over a pressure range from 0.7 to 1.5 MPa (100 to 200 psi). These results indicate that these 1-3-0 composite prototypes achieved some isolation from the lateral hydrostatic pressure but their performance would not place them beyond other 1-3 type composites with foamed cavities. The polymer matrix and encapsulated air gaps helped to decouple the negative contribution from lateral directions and enhance the hydrostatic piezoelectric response of the composite as a whole but was not without its problems. The integrity of sealing was very important. The strength of the sealant layer, designed to maintain the air gap and decouple the contribution from lateral directions, played a critical role in this composite. Because of the difference in compliance between the PZT-5H and the polymer matrix at these pressures, the difference in strain caused significant shear stress to

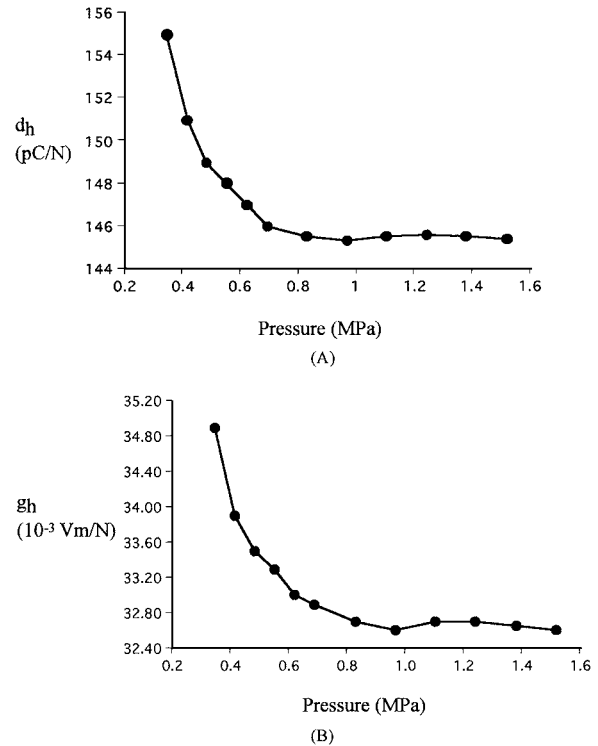


Fig. 3. Composite hydrostatic piezoelectric properties as a function of hydrostatic pressure. (A)  $d_h$  and (B)  $g_h$ .

develop and induced the rupture of the seal at the interface. Up to three broken air gaps were found in most samples which reduced the hydrostatic response of the composite. Further emphasis needs to be placed on the sealing methods and materials at the polymer and ceramic interfaces.

### Wiggle Hydrophone

A second type of SFF composite device was studied in order to try to improve the hydrostatic response of the 1-3-0 type hydrophone. We reasoned that if a contoured wall could be manufactured with the SFF technique we could redirect the lateral forces and improve upon the  $d_{33}$  response of the hydrophone. This was attempted with a modification of the 1-3-0 design which was made possible by the solid free-form method. In this design a curved-wall structure encloses the PZT-5H bar entirely within a smaller 'narrowed' air cavity. The main idea of the curved-wall structure is to aid in the mechanical amplification of the hydrostatic response of the enclosed piezoelectric element by creating tensional side wall stresses. The curved side-wall acts as a membrane

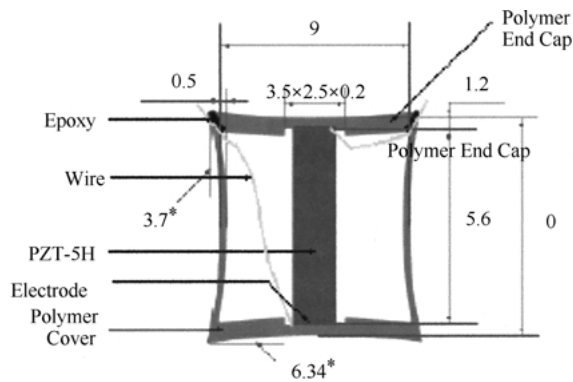


Fig. 4. Schematic of wiggle structure. units: mm.

under hydrostatic pressure and the displacement of the side-wall draws the two end caps towards each other, thereby inducing additional pressure on the PZT rod along its axial direction and amplifying the hydrostatic load. As a result, the overall hydrostatic piezoelectric response of the structure will be increased. The design requires that: (a) the top cap is very stiff so that the stress applied to the cap can be transferred effectively to the PZT element, (b) the side-wall is compliant so that it will not carry significant axial compression forces, and (c) the lateral pressure on the side-walls can be transferred to the PZT by tension forces. A schematic of the design is depicted in Fig. 4.

Because the PZT-5H bar and the curved wall are now acting to turn the negative piezoelectric contribution from the 1 and 2 directions into a positive contribution this encapsulation method may improve upon the original rigid wall design. Therefore, the hydrostatic coefficient  $d_h$  should approach that of the ideal  $d_{33}$  value.

The polymer walls (cover) and the end caps of the ‘wiggle’ hydrophone were designed and fabricated using the same stereolithography apparatus used in the straight wall design, the difference being the extremely thin sidewalls that were layered or ‘wiggled’ into position. Each layer was offset a small distance from the layer below to form the curved structure. In this way a thin almost membrane-like sidewall was built. Many iterations were required to develop a successful design. The SFF technique was an excellent tool for these intermediate design studies. This enabled us to optimize a final design to help increase the sensitivity.

The active ceramic elements were cut into the exact sizes for the fabrication process, two identical electric leads were attached to the electrode surfaces, with one hour required to cure the electrode epoxy in an oven

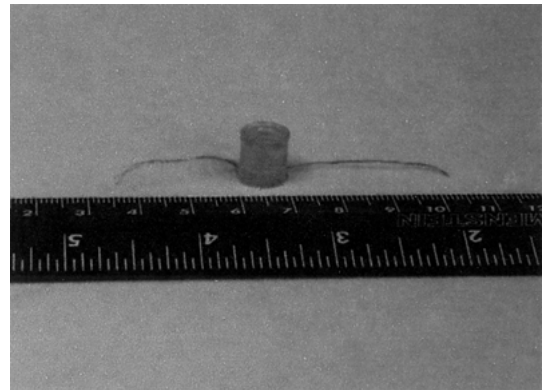


Fig. 5. Assembled wiggle hydrophone.

at 70°C. Thin layers of fast drying epoxy were applied onto the interfaces between the PZT bar, polymer cover, and the end cap in order to seal the device. To ensure the connectivity and encapsulation was successful, a 10 kg force was applied on both ends and held for 10 minutes. A schematic of the assembled ‘wiggle’ hydrophone is shown in Fig. 5.

The piezoelectric performance was evaluated with two PZT bar areas (cross sectional area) shown in Fig. 6 (A) and (B). The different size of the interface

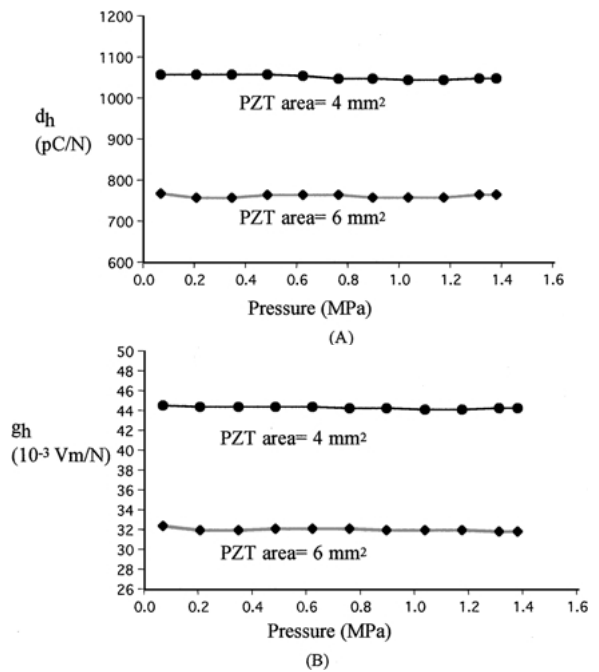


Fig. 6. “Wiggle” piezoelectric properties as a function of hydrostatic pressure. (A)  $d_h$  and (B)  $g_h$ .

between the PZT-5H bars and the polymer caps results in different pressures applied onto the bars. We tested four samples with a PZT area of  $\sim 4 \text{ mm}^2$ , and in the second we tested eight samples with an area of  $\sim 6 \text{ mm}^2$  each. The hydrostatic piezoelectric charge coefficient  $d_h$ , and the voltage coefficient  $g_h$  are plotted in Fig. 6(A) and (B) for each hydrophone element.

The samples showed stable performance under hydrostatic pressure, and no breakage was found after measurements. As shown in Fig. 6, for pressure up to 1.4 MPa (200 psi) the hydrostatic response is nearly constant, with the average  $d_h$  of the smaller bar ( $4 \text{ mm}^2$ ) yielding around 1055 pC/N, while that of larger area group ( $6 \text{ mm}^2$ ) was 752 pC/N. Comparing the  $d_h$  of the hydrophone to the  $d_{33}$  of a single PZT bar, the piezoelectric contribution from the lateral directions seems to be successively decoupled with further amplification of hydrostatic piezoelectric charge coefficient being achieved using this design technique. As expected, the smaller electrode area, yielded the higher piezoelectric response.

Finally, we must note that there are certain design and fabrication problems which must be controlled when building SFF encapsulated composites. Among the most critical are:

1. The structural alignment of the PZT within the encapsulation,
2. The lamination quality and surface curvatures, and
3. The tolerances of the walls and the endcaps especially near their interface and with respect to the ceramic.

## Conclusion

Compared to other hydrophone designs the encapsulation of 1-3-0 and wiggle hydrophones via SFF shows a wide range of hydrostatic response from 145 pC/N to 1055 pC/N depending on the specific geometry of the encapsulation. Of course, these performance figures are dependent on the size of the hydrophone and must be compared to similarly sized devices for determination of their relative performance and their possible applications. Surrounding air gaps in our initial straight walled design showed performance gain (145 pC/N) over the control sample (70 pC/N), but sealing problems would still need to be addressed. The structure of the wiggle hydrophone mechanically amplifies the hydrostatic piezoelectric response of PZT-5H by using

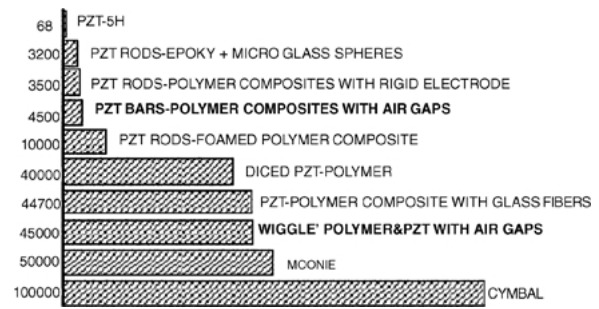


Fig. 7. Comparison of  $d_h g_h$  of various composites.

a complaint wall and shows the best performance improvement (1055 pC/N). In Fig. 7 we compare these and other composite hydrophone designs to demonstrate shows their approximate figures of merit [11].

Most composite devices are manufactured by more conventional forming methods. In this study, the Solid Freeform Fabrication technique was used to form dimensionally curved polymer parts, thereby, improving the composite design, saving time and minimizing human interference. This technique offers an approach where more complex design configurations may be attempted. The success of building simple designs with curved surfaces for amplified hydrostatic response shows that both the solid free-from method and computer-aided design can contribute to hydrophone technology.

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## References

1. O.B. Wilson, *An Introduction to Theory and Design of Sonar Transducers* (U.S. Government Printing Office, 1985).
2. R.E. Newnham, L.J. Bowen, K.A. Klicker, and L.E. Cross, *Mater. Eng.*, **2**, 93 (1980).
3. M.J. Huan, R.E. Newnham, and W.A. Schulze, *Advanced Ceram. Mater.*, **1**, 361 (1986).
4. J.F. Fernandez, A. Dogan, Q.M. Zhang, and R.E. Newnham, in *Fourth Euro-Ceramics*, Vol. 5, edited by G. Gusmano and E. Traversa (Electroceramics, Italy, 1995), p. 39.

5. R. Meyer, Jr., H. Weitzing, Q. Xu, Q. Zan, and R.E. Newnham, *J. Am. Ceram. Soc.*, **77**, 1669 (1994).
6. Q.M. Zhang, H. Wang, and L.E. Cross, *J. Mater. Soc.*, **28**, 3962 (1993).
7. A. Dogan, J.F. Fernandez, K. Uchino, and R.E. Newnham, *IEEE*, **1**, 213 (1996).
8. K. Onitsuka, Ph.D. Thesis, Pennsylvania State University, University Park, PA, 1993.
9. P.F. Jacobs, *Rapid Prototyping & Manufacturing: Fundamentals of Stereolithography* (Society of Manufacturing Engineers, Dearborn, 1992).
10. R.E. Newnham, L.J. Bowen, K.A. Klicker, and L.E. Cross, *Mater. Eng.*, **2**, 93 (1980).
11. L.E. Cross, in *Ferroelectric Ceramics: Tutorial Reviews, Theory, Processing, and Applications*, edited by N. Setter and E.L. Colla (Basel, 1993), p. 1.