Piezoelectric hydrophones from Optimal Design, II: properties

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Abstract Three-dimensional piezoelectric PZT-air artificial materials, designed using the Optimal Design by the Homogenization Method, with predicted hydrostatic piezoelectric coefficient of 427 pC/N and a hydrophone figure of merit of 29 pm²/N were realized and measured. The measured hydrostatic piezoelectric coefficient was 329 pC/N and a hydrophone figure of merit was19 pm²/N. The differences between predicted and observed properties were ascribed to insufficient polarization, due to the field distribution during poling.

Introduction

The general concept of creating designed structures with specific properties is familiar for macroscopic structures, such as vehicles or buildings. Usually designers optimize for such goals as maximum strength for minimum material. More recently, Optimal Design techniques have used computational Optimal Design methods to address other properties. The Topology Optimization and Homogenization algorithm of Bendsøe and Kikuchi [1] has been used to design the fine-scale structure for composite materials with

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F. M. de Espinosa Instituto de Acústica, Consejo Superior de Investigationes Científicas, Madrid, Spain pre-determined properties or properties not available in common materials. Such a fine-scale composite structure can be considered an "Artificial Material". This paper addresses piezoelectric artificial materials optimized for sensitivity as hydrophones, made from a microconfigured composite design of lead zirconate titanate (PZT) ceramic and air. The topology optimization and homogenization method used to design piezoelectric composites with elastic and piezoelectric coefficients optimized for transducer are presented in detail elsewhere [2–4]. Two-dimensional versions of these designs have been realized on a fine scale by Crumm et al. [5], who found excellent agreement of the predicted and measured properties. This paper concerns measurements on three-dimensional designs.

Silva's [3] designs were aimed at optimizing the performance of PZT as a hydrophone, so considered the hydrostatic piezoelectric response d_h (pC/N), which is the sum of the piezoelectric terms from all three directions

$$d_{\rm h} = d_{33} + d_{13} + d_{23} \tag{1}$$

Solid PZT, and otherwise excellent piezoelectric, is a poor hydrophone because the d_{33} coefficient has about the same magnitude, but opposite sign, as the sum of $(d_{13} + d_{23})$, so the d_h value is small. The hydrophone transducer sensitivity is determined by the hydrostatic voltage coefficient, g_h , which relates the measured voltage to an applied hydrostatic stress. The g_h and d_h terms are related by the relative permittivity of the material K and the permittivity of free space ε_o by:

$$g_{\rm h} = \frac{d_{\rm h}}{\varepsilon_o K} \tag{2}$$

Another useful quantity for a hydrophone transducer is the product of the hydrostatic piezoelectric response d_h and the

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hydrostatic voltage coefficient g_h , which is the figure of merit d_hg_h , has units of m²N⁻¹.

$$d_{\rm h}g_{\rm h} = \frac{d_{\rm h}^2}{\varepsilon_o K} \tag{1}$$

The hydrophone figure of merit can be increased by designing a transducer that either increases the hydrostatic piezoelectric response or decreases the permittivity. Silva's Optimal Designs combine voxels of air and PZT arranged to optimize one or both of these quantities. These designs are periodic structures with repeat units illustrated in Fig. 1. Design 2 was realized as a $3 \times 3 \times 1$ array of nine repeat units and Design 3 was realized as both a $3 \times 3 \times 1$ nine-unit array and a $10 \times 10 \times 10$ array of 1,000 repeat units. The fabrication methods used for realization artificial piezoelectrics as arrays of these repeat groups in PZT, along with images of the fabricated specimens, were presented in Part 1 of this series [6]. For comparison with the Optimally Designed materials, we prepared a $5 \times 5 \times 5$ array of 125 simple perforated PZT blocks. Figure 2 shows the simple design and the fabricated materials.

Hydrophone performance

Hydrostatic testing of the piezocomposites was performed at the Instituto de Acustica in Madrid, Spain. All of the fabricated hydrophone structures were poled in oil at 120 (C in a field in excess of 2 kV/mm. Samples were sealed with epoxy and placed under successive hydrostatic loadings of 5, 10, and 15 bar. The samples were pressurized under open circuit with the generated charge measured by a real time digitizing oscilloscope (Tektronics TDS520). The circuit was then closed to short out the stored charge. The pressure was relieved and the charge regenerated and recorded as the pressure dropped and stabilized. The charge was measured upon unloading as it yields a more stable result. The process was repeated three times at each pressure to ensure measurement repeatability.

Table 1 lists of the hydrostatic piezoelectric data for all samples. The piezoelectric charge coefficient (d_h) of the $3 \times 3 \times 1$ array of Design 2 was 208 pC/N compared to a predicted d_h value of 427 pC/N. While this is more than twice as sensitive as the solid PZT (100 pC/N), the fabricated Design 2 array is less than half of the predicted value for the $3 \times 3 \times 1$ array. The $d_{\rm h}$ for the $10 \times 10 \times 10$ array of Design 3 was 230 pC/N compared to a predicted value of 414 pC/N. For the $3 \times 3 \times 1$ array of Design 3, the piezoelectric charge coefficient d_h for Design 3 was 270 pC/N compared to a prediction of 414 pC/N. This is a 2.7× improvement over solid PZT, but still the measured hydrostatic charge coefficient was 65% of what was predicted. The hydrophone figure-of-merit d_hg_h for Design 3 was measured to be19 pm²/N. This should be compared to the very low d_hg_h for solid PZT, which is only 0.2 pm²/N. However, the predicted value for this design was $29.4 \text{ pm}^2/$ N. While this still represents a 2.7× improvement in $d_{\rm h}$ and an 95× improvement in d_hg_h over the PZT586 in the case of the $3 \times 3 \times 1$ array of Design 3, the measured performance as judged by both the d_h and d_hg_h hydrophone metrics are only 66% of expected performance for this design.

The simple perforated block, with voids having circular cross section, has the same connectivity and relative





Fig. 2 CAD file of the unit cell of the simple perforated design is a 3–2 pzt-air composite where the 2-d connectivity air filled channels has circular cross section. Also shown are a CAD file of a $5 \times 5 \times 5$ array of unit cells, the $5 \times 5 \times 5$ array fabricated in PZT by ISFF and an SEM of the unit cell faces

amounts void as of optimized Design 2 and Design 3 piezocomposite structures. This sample was measured under the same conditions, although the smaller size of these samples permitted a higher poling field (3 kV/mm).

Table 1 Hydrostatic piezoelectric performance of designs

| Material | Predicted hydrostatic charge coef. d _h pC/N | Measured hydrostatic charge coef. d _h pC/N | Predicted figure-of- merit $d_hg_h \text{ pm}^2/$ N | Measured figure-of- merit $d_hg_h \text{ pm}^2/$ N |
|---|--|---|---|--|
| Design $2 \ 3 \times 3 \times 1$ array | 427 | 208 | - | - |
| Design 3 $3 \times 3 \times 1$ array | 412 | 270 | 29 | 19 |
| Design 3 $10 \times 10 \times 10$ array | 412 | 230 | - | - |
| Simple design $5 \times 5 \times 5$ array | - | 166 | - | 4.5 |
| Solid PZT | | 100 | | 0.2 |
| Kumar et al. [8]–50% pores | | 58 | | 3.3 |
| Kara et al. [9]–50% pores | | 230–280 | | 14–16 |
| Bowen et al. [10]–50% pores | | | | 10–30 |

The measured d_h for the simplified structure was 166 pC/N compared to the optimized Design 3 structure with $d_h = 270$ pC/N. The measured d_hg_h for the simplified structure was 4.45 pm²/N compared to the optimized Design 3 with $d_hg_h = 19$ pm²/N. Both the optimized design and the simplified version offer significant performance advantages over the bulk properties of the PZT586.

It is interesting to compare the performance of these PZT-air composites having voids of specific designs, with PZT composites with random voids at a similar volume fraction. These are in the class of 3–3 piezocomposites [7], where both the solid and the void phase are have connectivity in all three-dimensions. Kumar et al. [8] reported on PZT with c. 100 µm spherical voids. Table 1 lists their data for PZT with about 50 volume percent voids, where $d_{\rm h}$ is 58 pC/N (compared to only 5 pC/N for their dense PZT), an enhancement of almost 12-fold. Their d_hg_h figure of merit was reported to be 3.3 pm²/N, compared to 0.02 reported for their solid PZT. Kara et al. [9] reported values for a wide range of PZT composites. Table 1 includes the range of values for samples around 50% void, with d_h is 230-280 pC/N (compared to only 72.8 pC/N for dense PZT), an enhancement of almost 4-fold. The d_hg_h values were in the range 14-16 pm²/N, compared to 0.37 pm²/N reported for their solid PZT. Bowen et al. [10] also report d_hg_h values ranging from 10 to 30 pm²/N for 50% porous 3-3 composites fabricated from PZT where the dense material had a figure of merit of 0.08 pm²/N. Thus the enhancement of hydrophone properties for the 3-3 PZT-air composites with random voids are comparable to materials with the designed microstructures.

The Optimal Designs indeed are superior to the simple perforated design, and much better than solid PZT, but the performance is only about 66% of the predicted performance. This is surprising in view of the must better agreement found by Crumm et al. [5] for two-dimensional designs. The two-dimensional designs were easier to fabricate, and were prepared as arrays with many more repeat units. The most likely causes for the discrepancy between the theoretical and experimental behavior is the state of polarization of the piezoelectric device, and the accuracy with which the optimized design was realized in the fabricated PZT. An analysis of the accuracy of these samples is reported in a companion paper [6], which has details on the Indirect Solid Freeform Fabrication method, shows that the actual PZT part dimensions vary from the design with a variance of (3%, depending on the design.

In the case of Design 2, the part fidelity was quite good when compared to the CAD file from which it was built. However, the CAD solid body model had vertical sections that were thicker than required by the voxelated FEM output from the Topology Optimization design scheme. This made the fabricated structure stiffer than the designed structure. A stiffer structure would have a lower measured hydrostatic charge coefficient, so some of the difference between predicted and measured could come from fabrication inaccuracies.

The properties in the as-fabricated PZT also could be different than the properties assumed in the design. In the Optimal Design procedure, the properties of the PZT are presumed to be the same as a well-poled dense material, and these are modified as the design evolves by replacing voxels of PZT with voids. This results in a prediction of properties for a well-poled piezoelectrics with a specific design of voids. However, we fabricate samples having the same design of voids, but from sintered unpoled PZT. This is a significant difference, since we later attempt to pole a composite with a complex arrangement of high permittivity solid and low permittivity voids.

Poling, or alignment of the ferroelectric domains within the material, is achieved by subjecting the piezoceramic to a high electric field, typically 2–5 kV/mm, at an elevated temperature. The piezoelectric properties obtained in the final device are heavily dependent upon the degree of domain orientation achieved during poling. However, voltage difference across a complex composite of high permittivity PZT ceramic and low permittivity oil will not create a uniform electric field. Simulations of the electric field distributions for a repeat unit for Design 3 were performed with the electrostatics package of Maxwell High Frequency Structure Simulation Software (Ansoft Corp., Pittsburgh, PA). The software takes the inputs of the hydrophone geometry with $\varepsilon_r = 4100$ immersed in oil with $\varepsilon_r = 5$, in an external electric field of 2 kV/mm, and solves for the spatial distribution of the magnitude and direction of the electric field and electronic displacement inside the oil and PZT. In some regions, the electric field was more than 1000× greater in the low permittivity oil than in the high permittivity PZT. The field vectors in the PZY ceramic were not well aligned in the direction of polarization.

The value of interest to the performance of the piezoelectric is the fraction of field vector in the desired polarization (z) direction. Ideally, this would approach 100% but the complex geometry and high permittivity of the PZT bend the field lines as they pass between dielectric media. The Maxwell Electrostatics software was employed to estimate the degree of alignment of the electric displacement, by integrating the electric field over the volume of the ceramic. The volume averaged field vector in the polarization direction for Design 3 was calculated to be only 67%. The lack of field alignment in the intended direction means that no matter how strong the applied electric field, complete ferroelectric domain z-axis alignment will never be achieved. The partial state of polarization a within the Design 3 hydrophone structure is likely a major factor accounting for the difference between the predicted and measured device performance. Irrespective of the alignment of the polarization vectors in the dielectric, a sample that is larger in the dimension of polarization will experience a lower strength polarization field. This is most likely explanation for the performance difference between the nine-unit $3 \times 3 \times 1$ array and the larger 1,000-unit $10 \times 10 \times 10$ unit cell arrays of design 3. The $10 \times 10 \times 10$ structure is over 3 times thicker in the polarization direction and therefore the average poling field was only one third as strong.

The calculated percentage of field vector in the polarization direction will vary from design to design. The same boundary conditions apply, but the differences in geometry result in variances in the electric field vectors. It is likely that complete polarization is unobtainable in any of the optimized hydrophone structures, but the extent to which the alignment differs from the idealized case will vary from case to case.

Conclusions

Three-dimesional, optimally designed hydrophones were successfully fabricated in good quality PZT via suspension polymerization in a lost mold made by a rapid prototyping tool. The Optimally Designed structures provide a 2.7× improvement in hydrostatic charge coefficient d_h and an 85× improvement in hydrophone figure-of-merit d_hg_h compared the bulk PZT586 from which they are fabricated. However, the performance is only about 65% of their expected performance predicted for these designs. The differences between the predicted and observed piezoelectric properties are attributed to insufficient polarization due to the electric field distribution in these complex objects during poling.

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