

Naval Research Laboratory

Washington, DC 20375-5320



NRL/MR/6300--96-7847

# Piezoceramic-Polymer Composites for Underwater Transducer Applications

C. KIM  
M. KAHN  
D. LEWIS, III

*Mechanics of Materials Branch  
Materials Science and Technology Division*

June 28, 1996

19960718 100

DTIC QUALITY INSPECTED 8

Approved for public release; distribution unlimited.

# REPORT DOCUMENTATION PAGE

*Form Approved*  
OMB No. 0704-0188

Public reporting burden for this collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden, to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington, VA 22202-4302, and to the Office of Management and Budget, Paperwork Reduction Project (0704-0188), Washington, DC 20503.

|   |                                     |   |  |
|---|-------------------------------------|---|--|
| 1. AGENCY USE ONLY ( <i>Leave Blank</i> )   | 2. REPORT DATE<br><br>June 28, 1996 | 3. REPORT TYPE AND DATES COVERED<br><br>Final 10/1/93-9/30/94 |  |
| 4. TITLE AND SUBTITLE<br><br>Piezoceramic-Polymer Composites for Underwater Transducer Applications   |                                     |   | 5. FUNDING NUMBERS   |
| 6. AUTHOR(S)<br><br>C. Kim, M. Kahn, and D. Lewis   |                                     |   |  |
| 7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)<br><br>Naval Research Laboratory<br>Washington, DC 20375-5320  |                                     |   | 8. PERFORMING ORGANIZATION REPORT NUMBER<br><br>NRL/MR/6380--96-7847 |
| 9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)<br><br>Office of Naval Research<br>Arlington, VA 22217-5660   |                                     |   | 10. SPONSORING/MONITORING AGENCY REPORT NUMBER                       |
| 11. SUPPLEMENTARY NOTES   |                                     |   |  |
| 12a. DISTRIBUTION/AVAILABILITY STATEMENT<br><br>Approved for public release; distribution unlimited.  |                                     |   | 12b. DISTRIBUTION CODE   |
| 13. ABSTRACT ( <i>Maximum 200 words</i> )<br><br>A piezocomposite structure was prepared in a 1-1-3 configuration by casting a compressible epoxy between lead zirconate titanate (PZT) rods and a stiff polymer preform matrix. A corrugated polymer preform was used to provide structural stability under high hydrostatic pressures. The changes of piezoelectric properties with pressure and temperature were very small. To enhance the piezoelectric responsivity, the 1-1-3 composite is further modified to have PZT rods extended beyond the epoxy matrix. The pressure applied to the cover plate is then transferred only to PZT rods and the pressure on the PZT rods is amplified by the reciprocal of the PZT volume fraction of the composite. As a result, a figure of merit can be achieved to more than $10,000 \times 10^{-15}$ (m <sup>2</sup> /N). |                                     |   |  |
| 14. SUBJECT TERMS<br><br>Piezoelectric ceramics<br>Lead zirconate titanate (PZT) rods<br>Polymer matrix   |                                     |   | 15. NUMBER OF PAGES<br><br>30  |
| Piezocomposite<br>Transducer<br>Hydrostatic   |                                     |   | 16. PRICE CODE   |
| Piezoelectric coefficients<br>Figure of merit   |                                     |   | 17. SECURITY CLASSIFICATION OF REPORT<br><br>UNCLASSIFIED            |
| 18. SECURITY CLASSIFICATION OF THIS PAGE<br><br>UNCLASSIFIED  |                                     | 19. SECURITY CLASSIFICATION OF ABSTRACT<br><br>UNCLASSIFIED   |  |
| 20. LIMITATION OF ABSTRACT<br><br>UL  |                                     |   |  |

## CONTENTS

|  |    |
|--|----|
| 1. INTRODUCTION .....  | 1  |
| 2. EXPERIMENTAL PROCEDURES .....                                   | 3  |
| 2.1 Perform Design .....   | 3  |
| 2.2 Materials Selection .....                                      | 4  |
| 2.3 PZT Rod Preparation Process and Characterization .....         | 5  |
| 2.4 Preparation and Tests of 1-1-3 Piezocomposite Prototypes ..... | 6  |
| 2.5 Rod Assembly Fixture .....                                     | 7  |
| 3. RESULTS AND DISCUSSION .....                                    | 7  |
| 4. CONCLUSIONS .....   | 12 |
| 5. ACKNOWLEDGMENT .....  | 13 |
| 6. REFERENCES .....  | 13 |

## 1. INTRODUCTION

Lead zirconate titanate (PZT) ceramics have been used for underwater transducers. However, single phase PZT ceramic has limited hydrostatic performance due to the low hydrostatic piezoelectric charge and voltage coefficients,  $d_h$  and  $g_h$ , respectively. Although PZT with large piezoelectric charge coefficients  $d_{33}$  and  $d_{31}$ , has been developed, the  $d_h$  is still low because  $d_h$  is equal to  $d_{33} + 2d_{31}$  where  $d_{33}$  is opposite in sign to  $d_{31}$  and has a magnitude only slightly higher than twice the magnitude of  $d_{31}$ . The  $g_h (= d_h / \epsilon)$  value is also small because the permittivity  $\epsilon$  is high for monolithic PZT.

In order to improve the hydrostatic piezoelectric sensitivities, the effects of the intrinsic coupling of  $d_{33}$  and  $d_{31}$  in single phase PZT are alleviated by using multi-phase composite structures. The composite properties can be determined by the choice of components, their relative amounts and the manner in which they are interconnected. In the past 15 years, many piezoceramic-polymer composites have been developed with the goal of obtaining significant improvement of their hydrostatic piezoelectric properties. Among these, the 1-3 type composite, with PZT rods aligned in the poling direction and embedded in a polymer matrix, is well adapted for potential hydrophone applications. Most 1-3 piezocomposites consist of two components, i.e. rods of PZT ceramics held parallel by a passive polymer matrix. However, this two phase configuration still has a limited hydrostatic charge responsivity due to a considerable contribution of the lateral  $d_{31}$  coefficient, that counteracts much of the uniaxial  $d_{33}$  response.

The objective of this project was to develop a low cost fabrication technology for high-performance 1-3 type piezocomposites that can be used in large area underwater transducers. The techniques are focused on the optimization of the hydrostatic sensitivities for specific applications and on the provision for a low cost, easily automated assembly method, suitable for the production of large area hydrophone devices.

Specifically, in order to minimize or neutralize the detrimental negative lateral pressure contribution ( $-d_{31}$ ) in 1-3 composites, a third component was added, in the present work, to a conventional 1-3 composite. It encloses both the individual piezoceramic rods and the passive polymer interphase layer in compartments of a honeycomb structure. The continuous rigid honeycomb skeleton provides 3-dimensional reinforcement and the passive polymer interphase can consist, moreover, of a foamed, soft polymer, to reduce its Poisson's ratio, thus allowing the piezoceramic rods to expand and contract freely in response to axial pressure or to applied electric field changes. This multiple component composite can be identified as a 1-1-3 piezocomposite[1]. That is, each phase of the composite, the ceramic rods, the soft polymer interphase and the continuous rigid skeletal structure, is self-connected in 1, 1 and 3 dimensions respectively (or the connectivity is 1-1-0-3 when air bubbles are introduced into the soft polymer interphase). In one fabrication method for the 1-3 system, lateral reinforcements were introduced by laying up glass fibers in the polymer interphase[2]. This configuration helps in reducing the lateral compression to some extent, but the polymer matrix still remains 3-dimensionally interconnected. Because of that, when such a reinforced 1-3 composite is modified by introducing air-bubbles into the polymer, the bubbles can collapse under high static pressures and introduce an unwanted pressure dependence. In contrast, the present approach is one in which both the ceramic rod and the polymer interphase are isolated collectively into each compartment of the reinforcing and stiff honeycomb matrix structure. By appropriate selection of materials and design of the honeycomb structure, lateral hydrostatic compression can be effectively reduced to maximize the hydrostatic response of the 1-1-3 piezocomposite.

Concurrently, the  $d_{33}$  coefficient of the composite can be optimized by modification of the interface between the composite and a cover plate to be applied to the composite so as to provide a pressure amplification that is proportional to the reciprocal of the volume fraction of the PZT rods in the composite. This also requires

removing the top polymer layer, thus providing a direct contact between the PZT rods and the cover plate.

Finally, to prepare large area 1-1-3 piezo-composites, a low cost and easily automated rod assembly method has to be developed, including an appropriately designed preform and a rod loading scheme. The approach here is to place the PZT ceramic rods in a parallel manner into a stiff polymer preform that has a corrugated cross section. The preforms with emplaced rods are stacked together and a soft epoxy interlayer is cast between the rods and the stiff polymer preform. This loading scheme could be automated to improve the assembly efficiency and further reduce cost.

With these goals in mind, design and fabrication of 1-1-3 piezo-composites, evaluation of the prototypes, and feasibility studies of the technology for large area transducers were conducted. In this report, results including preform design, materials selection, PZT rod preparation process and characterization as well as fabrication of composites, test and evaluation of prototypes with various cover plate modifications, and the concept of a mechanized rod loading fixture are documented.

## 2. EXPERIMENTAL PROCEDURES

### 2.1 Preform Design

The initial task of this project was to design a stiff corrugated preform for the preparation of 1-1-3 composites. The corrugated preform is designed such that PZT rods can be enclosed by the preform structure with space for a soft epoxy interlayer, with the rods distributed uniformly for a range of rod volume fractions, and with the rods easily loaded onto the preform in a semi-automated fashion. As shown in Fig. 1(A), the preform has spaced grooves and ridges in its upper and lower surfaces. Each upper groove provides space for only one PZT rod. The trapezoidal grooves and ridges provide a 3-dimensionally reinforced honeycomb structure when assembled on top of each other and glued together. Fig.1(B) shows the cross section of a PZT rod loaded preform assembly filled with a

soft resin interlayer. This preform design then provides an array of PZT rods in a hexagonal arrangement.

The upper groove dimensions (see Figs. 2(A) & 3) in the preform were chosen to allow the use of a range of PZT rod diameters that give PZT volume fractions from 10 to 25% as determined on the basis of only one rod occupancy for each groove. Specifically a rod, which can be of any diameter between 0.08 and 0.13 cm, can be placed in each groove when the rods are loaded in the way described in section 2.5. The maximum PZT volume fraction is determined by the largest diameter rods that fit into a given groove space. The minimum volume fraction is determined by the smallest diameter rod that would occupy each groove singly; that is, when loading rods into the grooves in the 45 degree inclined preform (see Fig. 4), rods of diameter larger than 0.08 cm can only occupy a given groove space singly because subsequent rods have contact with the first one, thus making the contact tangent angle with respect to the preform length larger than 45 degrees forcing them to roll over and slide to a next empty groove. With a given preform dimension, the volume fraction of the PZT is then determined by selection of PZT rod sizes, for example, 15 vol. % PZT can be obtained using 0.098 cm diameter rods as shown in Fig. 2(B). However, if only one rod size is available, for example, a rod diameter 0.13 cm, a lower volume fraction can be obtained by including in the assembly non-piezoelectric dummy rods as shown in Fig. 2(C). Also, when using two different rod sizes, the net PZT volume fraction can also be adjusted by mixing rods of the two sizes (Fig. 2(D)). In the two latter cases, the PZT rod distribution becomes irregular and loses its periodicity. Note that a non-periodic array of PZT rods is potentially beneficial in attenuating transverse resonances, when transverse resonance frequency waves are present and detrimental to performance by providing unwanted transverse coupling.

## 2.2 Materials Selection

Extruded PZT-5H rods were used in the preparation of the 1-1-3 prototype piezocomposites. These PZT-5H rods have high piezoelectric coefficients. However, their maximum allowable

compressive stress is low because depoling may occur under high longitudinal pressure. In that case, other types of PZT rods, such as PZT-4, may be substituted. A Rho-C epoxy is used as a compliant interphase layer, as this material has a very high mechanical compliance (Young's modulus 300 psi). Conathane EN-2 is another candidate for interphase layer and encapsulation material. For the preform, a stiff polymer is desired. Among many candidate polymers, polycarbonate such as Lexan is the first choice because of its superior mechanical properties and ease of moldability. Candidate material properties are given in Table 1.

### 2.3 PZT Rod Preparation Process and Characterization

PZT-5H rods have been characterized for their sintering and poling behavior. Early sample rods were prepared from extruded lengths of green rods provided by CPS and Cera Nova. These rods were cut and placed into a grooved zirconia slab and held in place by zirconia combustion boat covers. A vapor source was placed on an ungrooved slab. All slabs were loaded into a large boat and sealed with a ground cover. After these rods went through the usual burn out, sintering, and poling processes, the dielectric and piezoelectric properties were measured. XRF measurements showed that low  $d_{33}$  value rods (below 100 pC/N) have low Pb/Zr ratios (0.23-0.41:1) while the high  $d_{33}$  samples (above 450 pC/N) have high Pb/Zr ratios (approximately 1.20:1). This lead loss in the low  $d_{33}$  samples was remedied by seasoning the boats with PZ powder and physically covering the rods with a mix of PZT and  $ZrO_2$  powder in addition to having a PZ vapor source. Specifically, the burn out and sintering conditions were as follows: the organic binder was burned out in uncovered boat at 400°C for 29 hours and at 600 °C for 8.5 hours, and the samples were then covered and sintered at 1300 °C for 2 hours. For evaluating the  $d_{33}$  values of samples, short pieces of PZT rods were poled at 2.4 KV/mm for 10 minutes in 120 °C peanut oil.  $d_{33}$  values near 600 pC/N resulted with the processes given. PZT rods processed as shown were used in the preparation of a 1-1-3 prototype composite sample. Also square PZT rods supplied by AVX were processed similarly, for another prototype composite sample.

Results of processed and characterized PZT-5H rods received from various sources are given in Table 2.

#### 2.4 Preparation and Tests of 1-1-3 Piezocomposite Prototypes

A high stiffness Lexan plate was machined to have grooves and ridges suitable for a 17 vol % PZT composite with rod diameter 0.08 cm. For initial manual assembly the machined preform pieces were stacked together and assembled into a 2.2 x 1.6 x 0.8 cm honeycomb structure. The PZT rods (Cera Nova-1) were inserted into the holes of the preform skeleton. The rods and preforms were held in place by the low modulus, soft Rho-C epoxy. This epoxy was vacuum infiltrated into the assembly and cured for 12 hours at 70 °C.

The rod end surfaces of the assembled 1-1-3 composites were polished such that cover plates could be placed in direct contact with the rods. Grinding of the ceramic rods proceeds faster than that of the epoxy, leaving the epoxy slightly above the level of the ceramic. This happens because the soft epoxy is resilient and flexes around the polishing particles. Attempts were made to polish the epoxy level with the ceramic rods, including cryogenic polishing, use of soft media, wet polishing and chemical solvent polishing. None of these produced improved results. Therefore, a new approach to achieve a direct contact between the cover plate and the PZT rods was formulated, i.e., a modification of the cover plate configuration and of the composite surface as described in the section 3.

For the prototype 1-1-3 piezocomposite fabrication, injection molded preforms (Fig. 3) of plain polycarbonate Lexan and AVX PZT-5H square rods (0.1 x 0.1 cm) were used along with the choice of a soft epoxy interphase material. (However, the injection molded preforms can be used for 10 to 25 vol. % of PZT rods with diameter from 0.08 to 0.13 cm as shown in Figs. 2(A) & 3.) Thus prepared composites were encapsulated with Lexan edge strips and electroded cover plates, and then Corona poled. The Corona discharge poling processes were conducted at 60 °C under 10 KV for 10 min. The hydrostatic sensitivity measurements were conducted at NRL-USRD.

## 2.5 Rod Assembly Fixture

The assembly of PZT rods into the corrugated preforms was demonstrated by building a large (four-times) model fixture as shown in Fig. 4. The rods are initially aligned parallel to each other by shaking and vibrating the loading fixture. This rod loading fixture is applied normal to the model preform, which is inclined about 45 degrees from the horizontal plane, such that they form a V shape. By sliding the rod loading fixture along the preform surface, the rods are automatically deposited into the grooves. The grooves are designed to hold only one rod each as described in Section 2.1. The essential requirement of the loading unit is that the rod loader's bottom plate edge be aligned parallel to the groove length, such that rods can be placed into the grooves smoothly. This can be easily achieved by using extended guide plates in the loader to fit with the width of the preform. The model fixture is being used to evaluate the feasibility of the automated (actual size) loading fixture.

## 3. RESULTS AND DISCUSSION

The first prototype 1-1-3 piezocomposite was tested for its dielectric and piezoelectric properties which are given in Table 3. This sample contained 17% of PZT rods with a rod diameter of 0.08 cm, 53% of Lexan preform and 30% of a Rho-C soft epoxy interphase layer. The sample surfaces were polished with SiC papers as smooth as possible. However, as mentioned above, the resilient soft epoxy remained extended above the polished ceramic surfaces. One side of the sample surface was then electroded with a high viscosity Ag paste (0.1 cm thick) and the opposite end surface was electroded with a low viscosity Ag paint. The hydrostatic strain coefficient,  $g_h$ , was measured under ambient pressure using a reciprocity method and the value was  $44 \times 10^{-3}$  Vm/N. The figure of merit,  $g_h \times d_h$ , was about  $6,000 \times 10^{-15}$  m<sup>2</sup>/N, where that of the glass fiber reinforced diced composite shown in ref.2 was  $3,264 \times 10^{-15}$  m<sup>2</sup>/N. When measuring  $d_{33}$  with a Berlincourt meter, the values obtained depend upon the test probe used and on their position on the composite surface.

The piezoelectric constant  $d_{33}$  measured at constant electric field is defined as follows:

$$d_{33} = \left( \frac{\partial D_3}{\partial \sigma_3} \right)_E \quad (1)$$

where  $D$  and  $\sigma$  represent charge density ( $C/m^2$ ) and mechanical stress ( $N/m^2$ ), respectively. Expression (1) can be simplified to:

$$d_{33} = \frac{Q/A}{F/A} = \frac{Q}{F} \quad (2)$$

where  $A$  is the area stressed by the force  $F$  and, here, it is the same as the electroded area. The  $d_{33}$  value is then independent of area for homogeneous PZT. However the 1-1-3 composite surface is inhomogeneous. The  $d_{33}$  values therefore depend on whether the test probe is in physical contact just with the PZT rods or also with the polymer matrix. The  $d_{33}$  value also depends on the type of probe used, i.e., point, wedge or flat probes, as furnished with the Berlincourt  $d_{33}$  meter. Therefore, the behavior of the stress response was investigated with differently shaped probes applied on the top surface of the composite. Alternately various sizes of Ag paint-coated alumina plates were used as the testing probes, by inserting them in between the sample and the regular Berlincourt  $d_{33}$  meter wedge probe. The probe used for the bottom surface of the sample was a flat-surfaced, round ram (dia. 1.23 cm). The recorded  $d_{33}$  values were each averaged from at least ten measurements and are plotted as a function of the flat alumina plate area as shown in Fig. 5a. The best fit curve shows the  $d_{33}$  values decrease proportionally to the logarithm of the plate area. This indicates that the  $d_{33}$  in a 1-1-3 composite is strongly dependent on the area of the test probe, not as shown in equation (2) that applies only to homogeneous materials. When the alumina test plate was modified to have a corrugated surface, in which the ridges are aligned with the arrays of ceramic rod in the composite, a similar area dependency was also observed as shown in Fig. 5b, but this time the  $d_{33}$  values are consistently higher than when the flat plates were used. From this,

one would expect a similar response improvement in hydrostatic tests, as long as a seal is provided around the edge of the cover plate.

These observations lead to the preparation of modified cover plates designed to maximize the piezoelectric sensitivity of a 1-1-3 piezocomposite. Specifically they are modified from a flat alumina plate (Fig. 6a) to have a corrugated (Fig. 6b & 6c) or a cross-hatched surface (Fig. 6d), where the columns of ridges or arrays of bumps are located opposite the arrays of PZT rod in the composite. Such cover plates are then used on the polished surface of a 1-1-3 composite with a 17 vol % of PZT rods. The dimensions of the composite and the cover plates are 2.12 cm x 1.57 cm and 2.025 x 1.55 cm respectively. Thus the cover plate covers most of the composite area. The effects of each cover plate were tested one at a time on the same 1-1-3 composite by measuring the piezoelectric charge coefficient,  $d_{33}$ , with the Berlincourt  $d_{33}$  meter. The average  $d_{33}$  values of at least ten measurements, each using one of the three different cover plate modifications, are  $206 \pm 15$ ,  $344 \pm 14$  and  $405 \pm 7$  pC/N as given in Table 3, while the nominal contact area percentiles of the cover plate with the polymer matrix are 80, 30 and 7, assuming optimum alignment, using flat, corrugated and cross-hatched cover plates, respectively. The results are plotted in Fig. 7a. With the latter two cover plate modifications, the measured  $d_{33}$  values increased by 67% and 97% respectively over those measured with the flat plates. This is because they have a reduced contact area with the polymer matrix while maintaining optimum contact with the PZT rods. Although the corrugated cover plate may still have about 30% contact area with the matrix, it provides an enhancement of the piezoelectric response by more than 60%. The corrugated cover plate can be easily mounted and aligned with the arrays of PZT rods in the composite. The cross-hatched cover plate improves the sensitivity further but it needs more careful alignment for a good matching contact between the high spots (bumps) on the plate and the PZT rods.

In order to determine the effect of the Ag paste electrode at the bottom surface of the specimen, the bottom electrode was

replaced with a thin Ag paint-coated alumina plate which was bonded to the composite with a layer of Ag paste. Use of the alumina plate raised all the measured  $d_{33}$  responses by about 165 pC/N. The  $d_{33}$  values still show a similar contact area dependency using different types of top cover plates (Fig. 7b). It is interesting to note that upon extrapolation of  $d_{33}$  to zero contact area with the matrix, its value is near 600 pC/N. This corresponds to the average  $d_{33}$  value of individual PZT rods. The lower  $d_{33}$  values measured when there was only a Ag paste electrode on the bottom may be due to relaxation of stresses in the paste bottom layer.

There may be a possible additional mechanism to explain why the modified cover plates yield higher stress sensitivity. Although the interphase Rho-C epoxy is soft and highly flexible, the epoxy, when completely enclosed in the stiff honeycomb structure, is not easily deformable because of low volume compressibility. The volume compressibility is the reciprocal of the bulk modulus,  $B$ , defined as

$$B = Y / [3 (1-2 \nu)] \quad (3)$$

where  $Y$  and  $\nu$  are the Young's modulus and the Poisson's ratio respectively. Materials with a Poisson's ratio of 0.5 are incompressible because, as equation (3) shows, the bulk modulus  $B$  becomes infinitely large, regardless of the value of  $Y$ . When the flat cover plate is used to transmit pressure to the 1-1-3 composite surface, the epoxy material extending over the rods can not be easily compressed into the rigid preform unless there is compressible space provided, such as air bubbles or macrovoids. On the other hand, the modified (corrugated and cross-hatched) cover plates provide open spaces into which the excess epoxy can be easily displaced and deformed, allowing the ridges and bumps of the cover plates to have a good direct contact with the PZT rods. A schematic drawing of a side view of a composite with a corrugated cover plate is given in Fig. 8a. When the 1-1-3 composite with the corrugated alumina cover plate was compressed under 2 MPa (290 psi) of hydrostatic pressure, the soft epoxy had a permanent deformation, showing protrusions of

the epoxy surface into the open grooves of the corrugated plate as shown in Fig. 8b.

This leads to another composite surface modification, i.e. the use of extended PZT rods. When a flat cover plate is used on a ground composite surface, the pressure applied to the cover plate,  $\sigma_{cp}$ , is distributed to the PZT rods and the polymer (p) matrix according to the following equation:

$$\sigma_{cp} = f_{pzt} \sigma_{pzt} + f_p \sigma_p \quad (4)$$

where  $f$  represents the volume fraction. A significant portion of the applied force may then be transferred to an electrically inactive polymer matrix. Only the remainder is applied to the PZT rods, which are the source for generating the piezoelectric response. Less amplification can then result, in particular with higher stiffnesses and a higher Poisson's ratio of the polymer matrix. An optimization of force transfer can be achieved by preparing an extended PZT rod composite and providing it with a flat cover plate. The pressure applied to the cover plate is then transferred to the PZT rods only [3]. Equation (4) then reduces to:

$$\sigma_{pzt} = \beta \sigma_{cp} \quad (5)$$

where  $\beta$  is a stress amplification factor equal to the reciprocal of the PZT volume fraction.

To demonstrate the effect of the extended PZT rod configuration, a prototype piezocomposite was prepared as shown in Fig. 9. The composite was made of AVX PZT-5H rods and polymer matrix elements. The matrix was consisted of rigid Lexan preforms and soft EN-2 epoxy interlayers. This composite was cut in into two parts, one with the extended rods (P10B) and the other with the plain composite surface (P10A). Thus, prepared samples were tested and the results of the hydrostatic coefficients,  $d_h$  and  $g_h$ , as a function of static pressures up to 7 MPa are shown in Fig. 10. It is clear that the extended PZT rod sample demonstrates an enhanced piezoelectric

performance. However, the piezoelectric properties are not quite proportional to the stress amplification as predicted in equation (5). A possible explanation of this discrepancy may be due to the fact that most of the PZT rods are embedded in the matrix and all the pressure applied to the extended rod cannot be transferred through the whole length of the rod because a fair portion of the stress will be absorbed by the matrix. The figure of merit was nevertheless increased to 11,000 from 5,800 ( $\times 10^{-15} \text{ m}^2/\text{N}$ ) by the extended PZT rod configuration. The pressure dependence of the hydrostatic coefficients of both samples are small but the extended rod configuration sample has slightly higher pressure dependence. The dependence of  $d_h$  and  $g_h$  values on temperature also shows a small variation as shown in Fig. 11. The  $d_h$  and  $g_h$  increase as the temperature increases from 0 to 20 °C. Here also the extended rod composite had enhanced  $d_h$  and  $g_h$  values although they had more sensitive variation with temperature below 20 °C. The temperature and pressure dependence of the dielectric constants,  $K_{33}$ , of both configurations are almost identical.

#### 4. CONCLUSIONS

High sensitivity 1-1-3 piezocomposite transducers were prepared using corrugated, stiff preforms and a compliant epoxy resin into which PZT-5H rods were embedded. A cost effective assembly method for the large area 1-1-3 composite array was demonstrated with an automatic rod loading mechanism that uses specifically designed corrugated preforms and loading fixtures. A transducer assembly, that had extended PZT rods with air cavities between the polymer matrix and the cover plate, demonstrated a figure of merit of more than  $10,000 \times 10^{-15} \text{ (m}^2/\text{N)}$  at 1 MPa, 1 KHz and 20 °C. The 1-1-3 prototype composites also demonstrated only a marginal change of piezoelectric properties with temperature and pressure.

## 5. ACKNOWLEDGEMENT

The authors wish to thank Kurt M. Rittenmyer for the hydrostatic coefficients measurements conducted at NRL-USRD, and Mark Chase at NRL for PZT rod preparation.

## 6. REFERENCES

- [1] M. Kahn and C. Kim, "Transducers and Method for Making Same", U.S. Patent No. 5,325,011, June 28, 1994.
- [2] N. M. Shorrocks, M. E. Brown, R. W. Whatmore, and F. W. Ainger, "Piezoelectric Composites for Underwater Transducers", *Ferroelectrics*, 54, 215, (1984).
- [3] C. Kim and M. Kahn, "Transducers with Improved Signal Transfer", U.S. Patent No. 5,376,859, Dec. 27, 1994.

Table 1. PREFORM AND EPOXY MATERIALS FOR 1-1-3 PIEZOCOMPOSITES

| Property                    | LEXAN<br>3413 | LEXAN<br>141 | RHO-C<br>35065 | HD-50   | HD-35   | CONATHANE<br>EN-2  |
|-----------------------------|---------------|--------------|----------------|---------|---------|--------------------|
| <u>GENERAL, @ 23°C</u>      |               |              |                |         |         |                    |
| Density (gr/cc)             | 1.43          | 1.2          | 0.94           | 1.053   | 1.043   | 1.04               |
| Filler Content, %           | 30            |              |                |         |         |                    |
| Water Absorption, 24hrs, %  | 0.14          | 0.15         | 13.6           | 9.7     | 0.40    |                    |
| Injection Molding           | yes           | yes          |                |         |         |                    |
| Cure Cycle, °C              |               |              | 24 hr@80       | 1 hr@70 | 2 hr@85 |                    |
| <u>MECHANICAL, @ 23 °C</u>  |               |              |                |         |         |                    |
| Tensile Strength, ksi       | 19            | 10           | 0.139          | 0.071   | 0.800   |                    |
| Tensile Modulus, ksi        | 1250          | 14           | 0.404          | 0.143   | 0.600*  |                    |
| Flextural Strength, ksi     | 23            |              | 0.3            |         |         |                    |
| Flextural Modulus, ksi      | 1100          | 340          |                |         |         |                    |
| Poisson's Ratio             |               |              | 0.18           | 0.20    | 0.43*   |                    |
| <u>ELECTRICAL, @ 23 °C</u>  |               |              |                |         |         |                    |
| Dielectric Strength, KV/mm  | 18.7          | 15.0         | 4.75           | 4.75    | 4.01    |                    |
| Dielectric Constant, 10 KHz | 3.35          | 3.17         | 0.065          | 0.029   | 0.038   |                    |
| Dissipation Factor, 1 MHz   | 0.007         | 0.010        |                |         |         |                    |
| <u>MATERIAL SOURCE</u>      | G.E.          | G.E.         | BF Goodrich    | FMI     | FMI     | CONAP<br>*MSI data |

Table 2. RESULTS OF PROCESSED PZT-5H RODS RECEIVED FROM VARIOUS SOURCES.

| SOURCE                    | CPS-1 | CPS-2 | CeraNova-1 | Cera Nova-2 | AVX-1       | AVX-2       | CPS-3 | CPS-4 |
|---------------------------|-------|-------|------------|-------------|-------------|-------------|-------|-------|
| LENGTH (cm)               | 0.3   | 0.17  | 0.29       | 0.16        |             |             | 0.43  | 0.37  |
| DIAMETER (cm)             | 0.07  | 0.07  | 0.07       | 0.07        | 0.07 X 0.07 | 0.07 X 0.07 | 0.114 | 0.11  |
| CAPACITANCE (pF)          | 3.488 | 5.876 | 3.115      | 4.602       |             |             | 6.54  |       |
| LOSS TANGENT              | 2.00% | 3.15% | 2.70%      | 2.83%       | 2.13        | 1.75        | 1.75  | 3.5   |
| d33 VALUE                 | 511   | 512   | 600        | 552         | 482         | 574         | 383   | 447   |
| DIELECTRIC CONSTANT, K    | 3072  | 2933  | 2652       | 2162        | 3799        | 3122        | 3112  | 2794  |
| POLING FIELD (kV/mm)      | 2.4   | 2.4   | 2.4        | 2.4         | 2.4         | 2.4         | 2.4   | 2.4   |
| POLING TEMP (°C)          | 130   | 130   | 130        | 130         | 120         | 120         | 120   | 120   |
| POLING CURRENT(microamps) | 6     | <6    | <5         | <6          |             |             |       |       |

- CPS Superconductor Corp., Mass.
- Cera Nova Corp., Hopkinton, Mass.
- AVX, S. Myrtle Beach, SC

Table 3. DIELECTRIC AND PIEZOELECTRIC PROPERTIES OF PROTOTYPE 1-1-3 COMPOSITES WITH VARIOUS COVER PLATES(CP)

| Electrode type:<br>Top Surface CP<br>(Bottom Surface) | $K_{33}$ | $d_{33}$ | $g_{33}$ | $g_h$ | $d_h$ | $g_h d_h$ | dis. F.<br>% |
|---|----------|----------|----------|-------|-------|-----------|--------------|
| (1) Ag paint<br>electrode only<br>(Thick Ag Paste)    | 340      | 320**    | 106      | 45    | 135   | 6040      | 2.5          |
| (2) Flat CP   | 406      | 206      | 57       |       |       |           | 2.14         |
| Corrugate CP  | 406      | 345      | 96       |       |       |           | 2.14         |
| Cross-Hatch CP<br>(Thick Ag Paste*)                   | 406      | 405      | 113      |       |       |           | 2.14         |
| (3) Flat CP   | 406      | 370      | 103      |       |       |           | 2.14         |
| Corrugate CP  | 406      | 524      | 146      |       |       |           | 2.14         |
| Cross-Hatch CP<br>(Ag paint coated<br>Alumina)        | 406      | 564      | 157      |       |       |           | 2.14         |

Units of  $d_{33}$ ,  $d_h$ , pC/N;  $g_{33}$ ,  $g_h$ ,  $10^{-3}$  Vm/N;  $g_h d_h$ ,  $10^{-15}$  m<sup>2</sup>/N

\* E-Solder 3021 Silver Epoxy, Insulating Materials Inc. ACME, New Haven, Conn.

\*\* Used round ram probe without any cover plates  
 $g_h$  was measured under ambient pressure at Naval Underwater Warfare Center, Mass.  
 $d_{33}$  was measured with a Berlincourt  $d_{33}$  meter, Model CPDT-3300, Channel Products Inc.

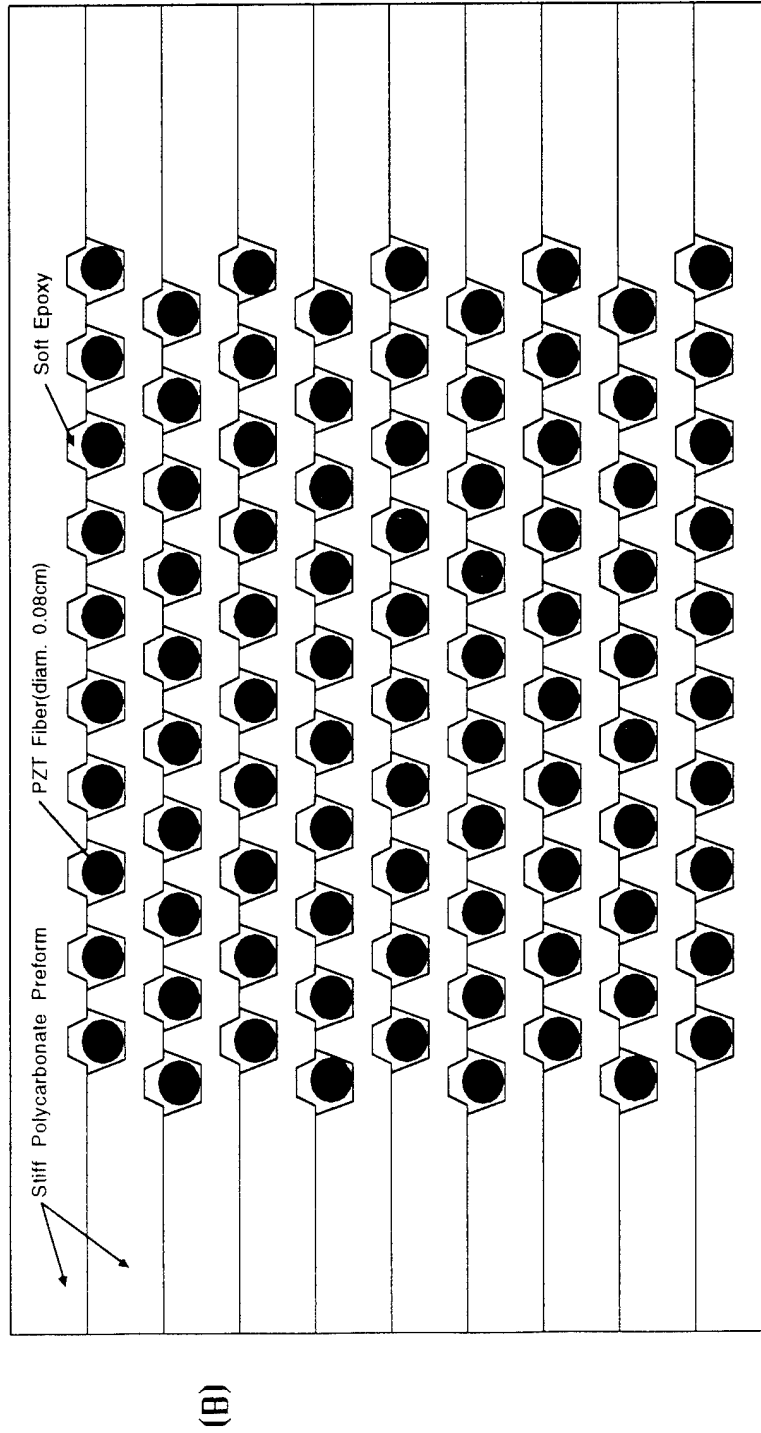
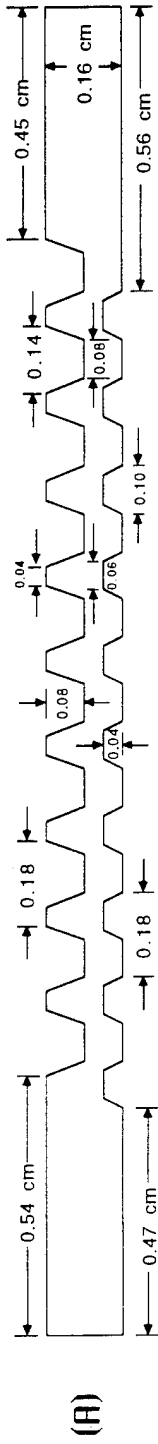


Fig. 1. (A) Preform Design and (B) Prototype 1-1-3 Piezocomposite Assembly for 17% PZT Fiber, 25% Epoxy & 58% Lexan Preform

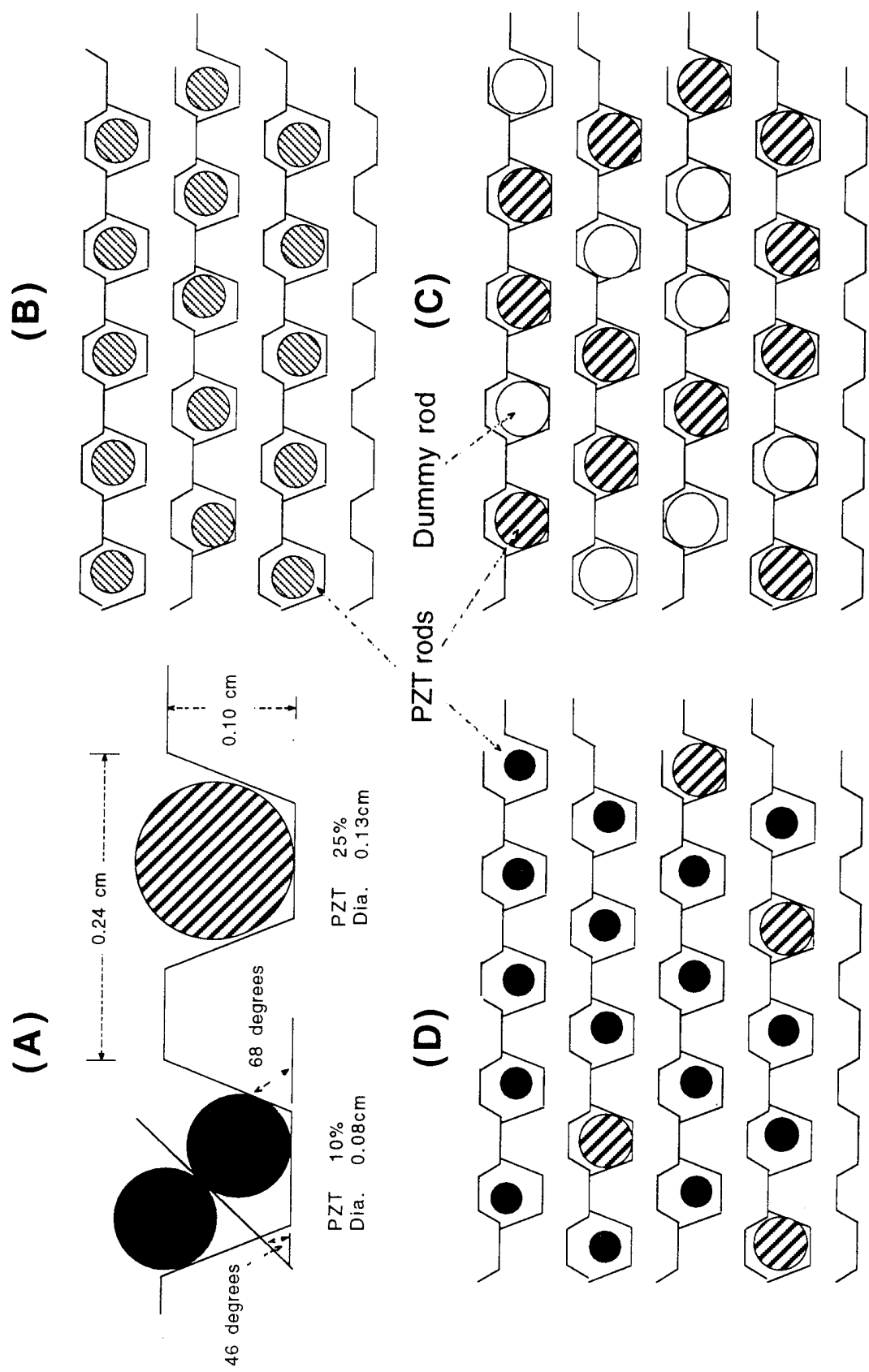
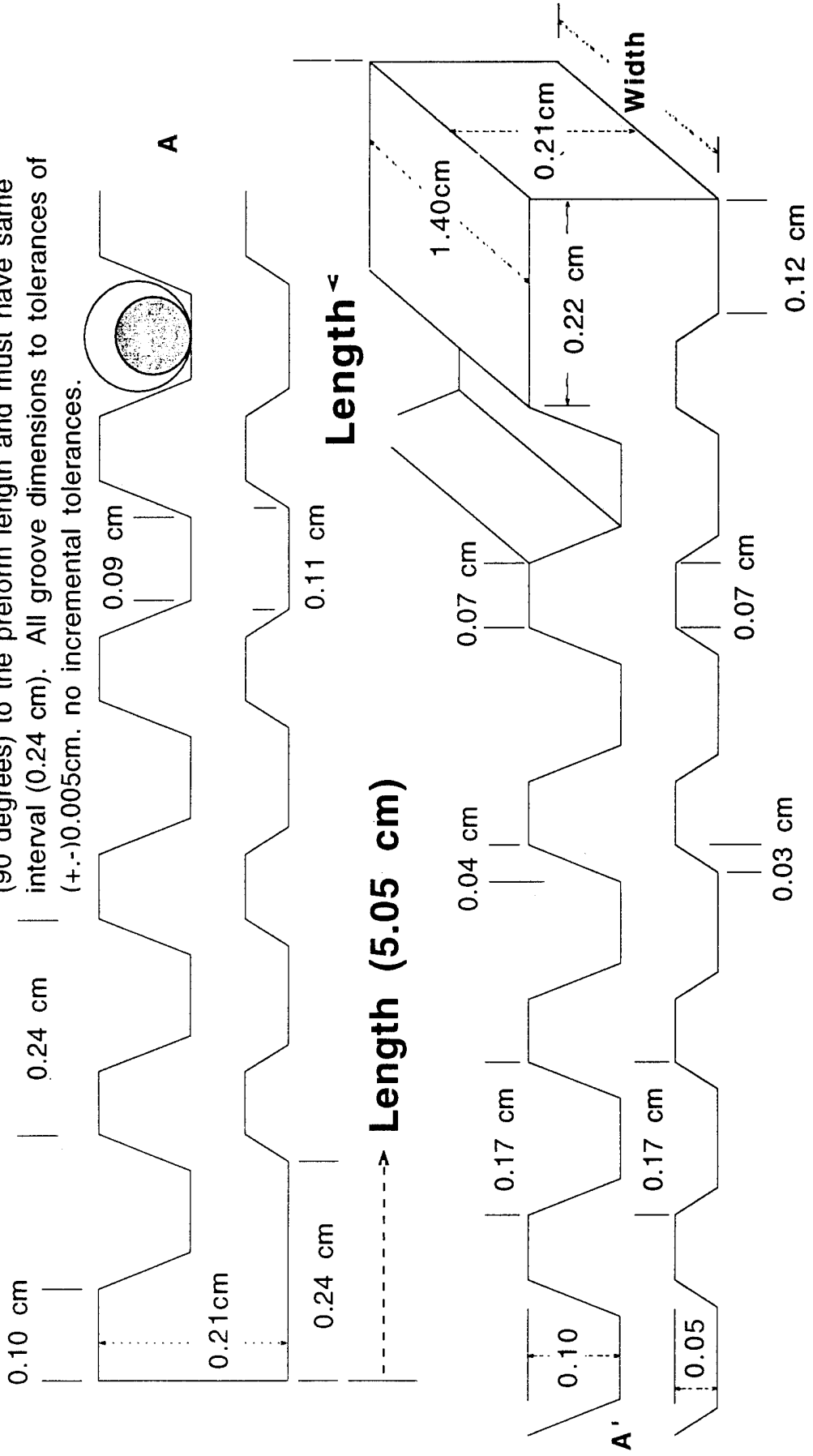


Fig. 2. (A) Range of PZT Vol % & Rod Diameters, (B) 15% PZT, Dia. 0.098cm, (C) 15% PZT, Dia. 0.13cm, with Dummy Rods, (D) 10% PZT, Mixed Dia. 0.13 cm & 0.08 cm.

**Fig. 3. Preform Design for Injection Molding.**

Total length; 5.05 cm, width; 1.40 cm, thickness; 0.21 cm  
 Number of grooves; total 20 each on top and bottom,  
 Material; Polycarbonate(Lexan or Hyzod),  
 A continues to A' with 10 more grooves between A and A' which are not shown here, the grooves must be normal (90 degrees) to the preform length and must have same interval (0.24 cm). All groove dimensions to tolerances of (+.-)0.005cm. no incremental tolerances.



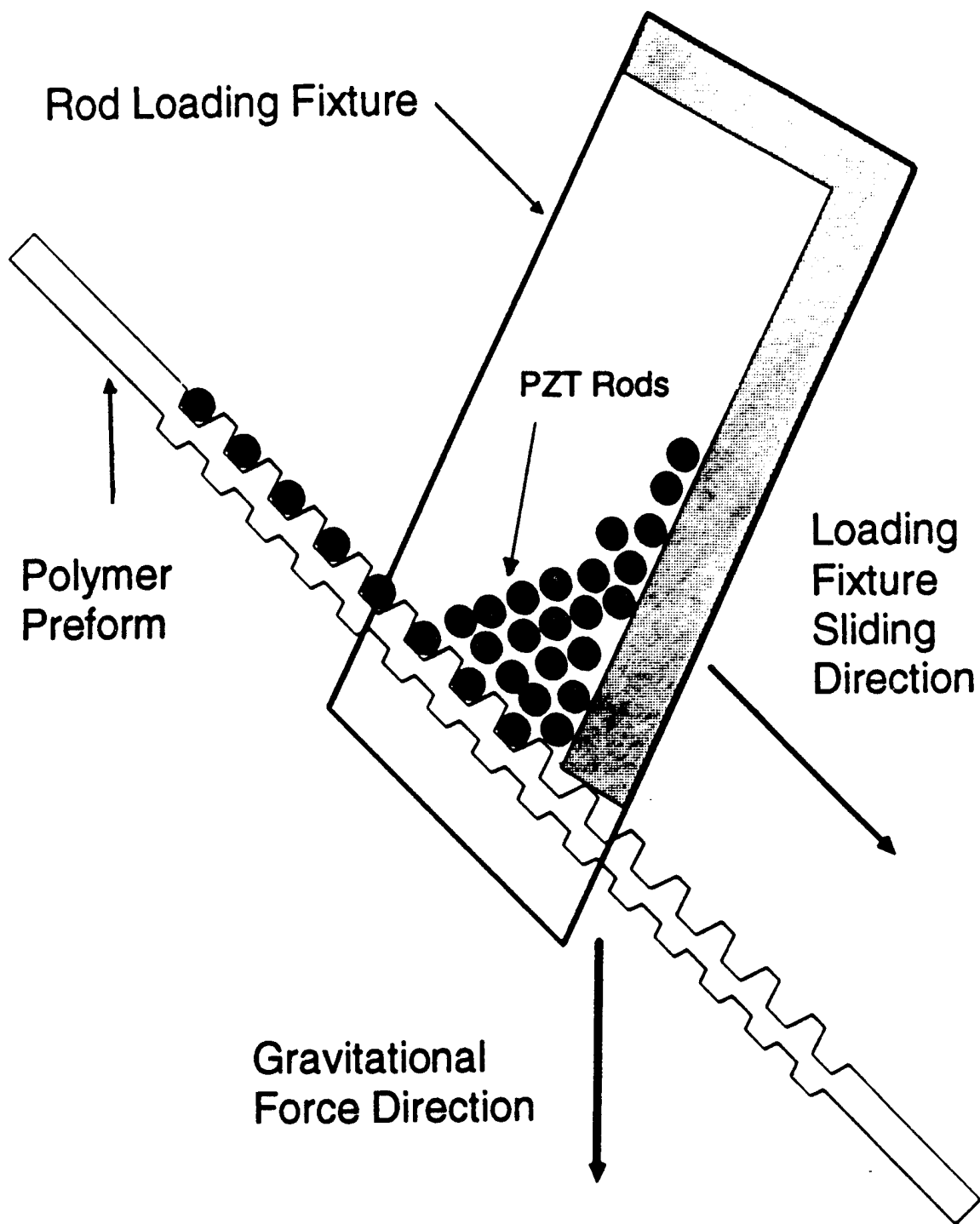


Fig. 4. Schematic of rod assembly fixture showing the loading mechanism

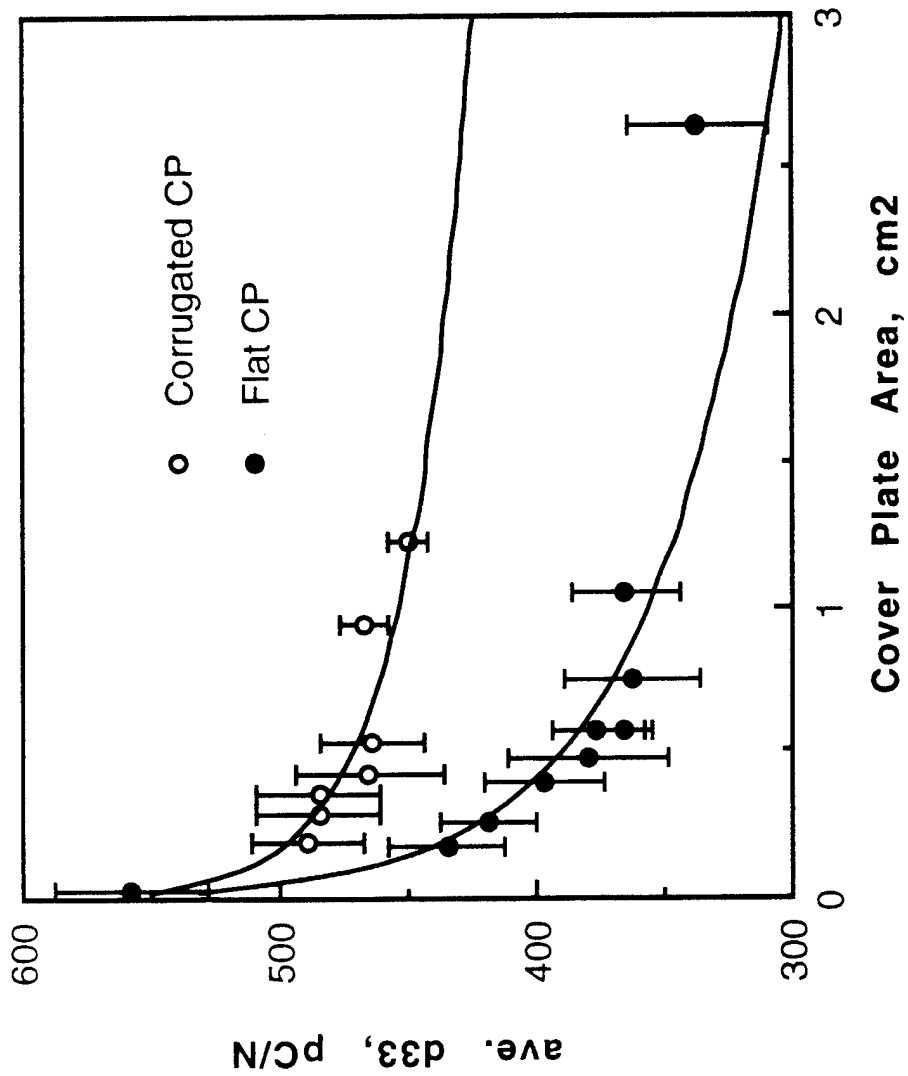


Fig. 5. Measured d33 values as a function of cover plate area, (a) ---●---Flat, (b) --○--Corrugate

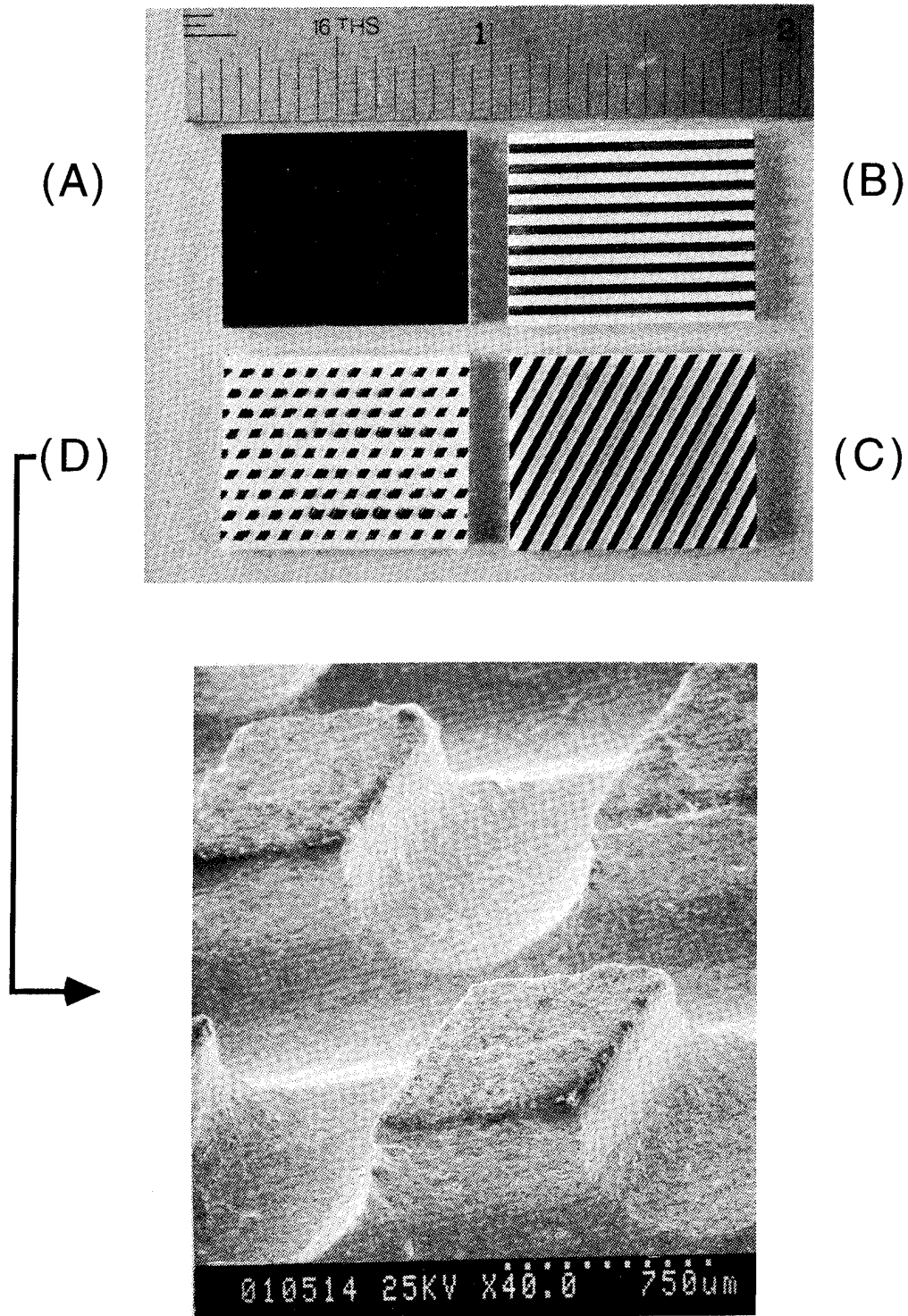


Fig. 6. Various cover plate configurations: (A) Flat, (B) & (C) Corrugated, (D) Cross-Hatched

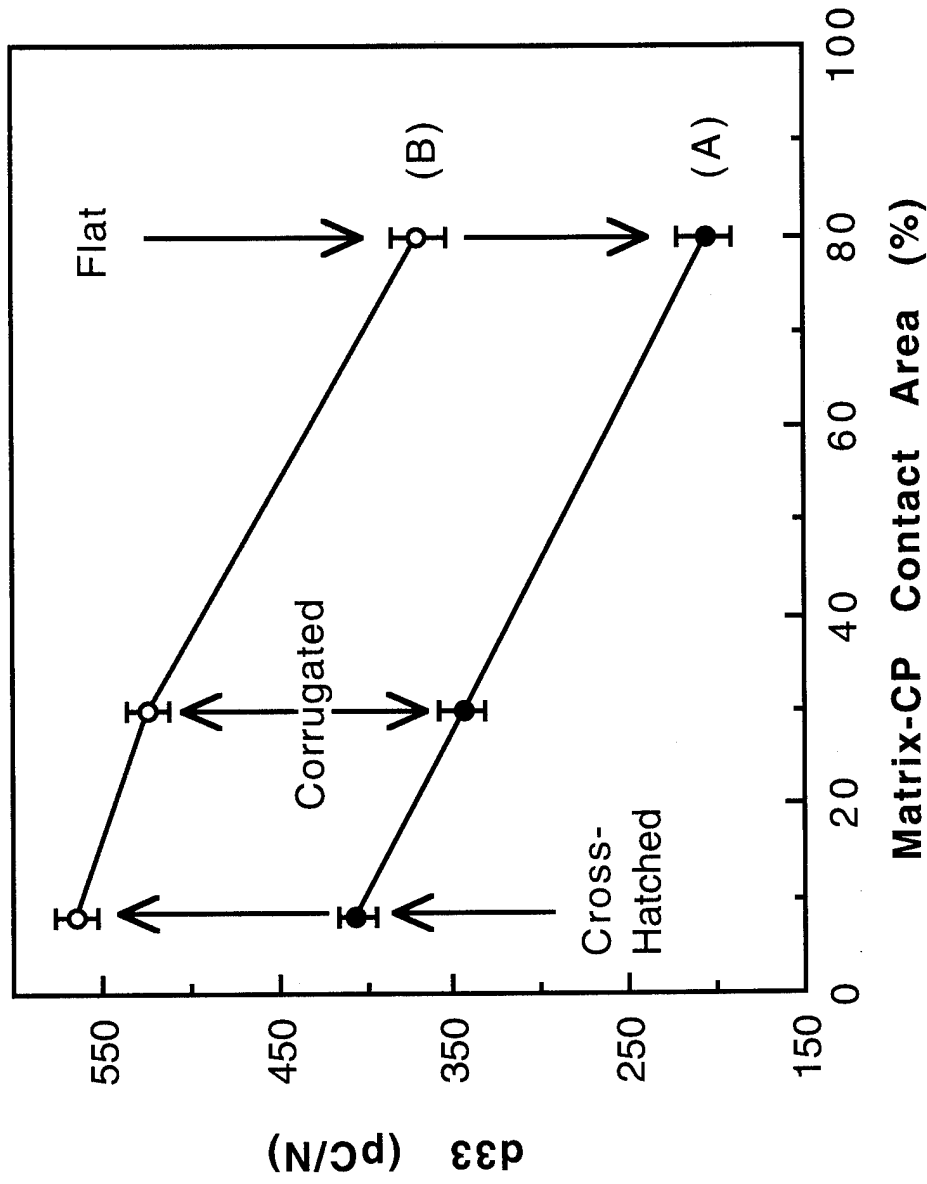
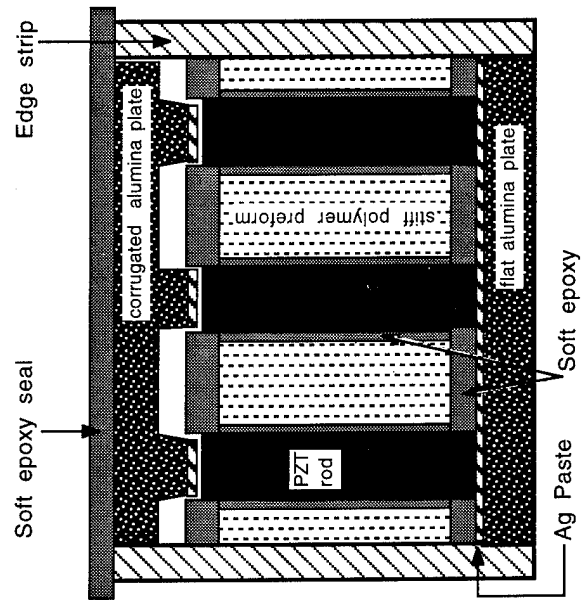
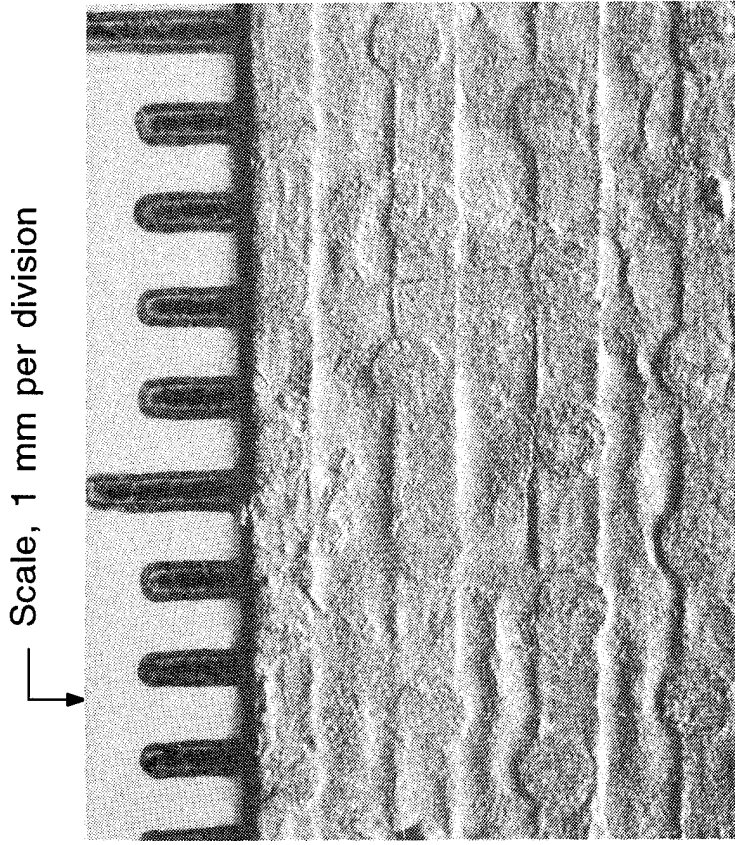


Fig. 7. Measured d33 values as a function of nominal matrix-cover plate contact areas for two different bottom surface electrodes, (A) Thick Ag paste, (B) Rigid alumina. Matrix area here includes both preform and epoxy areas.



(A)

(B)

Fig. 8. Prototype 1-1-3 composite for d33 and dh measurements, (A) Schematic of the composite, side view, (B) Sample top surface view of the impression made by arrays of grooves and ridges at 2MPa hydrostatic pressure.

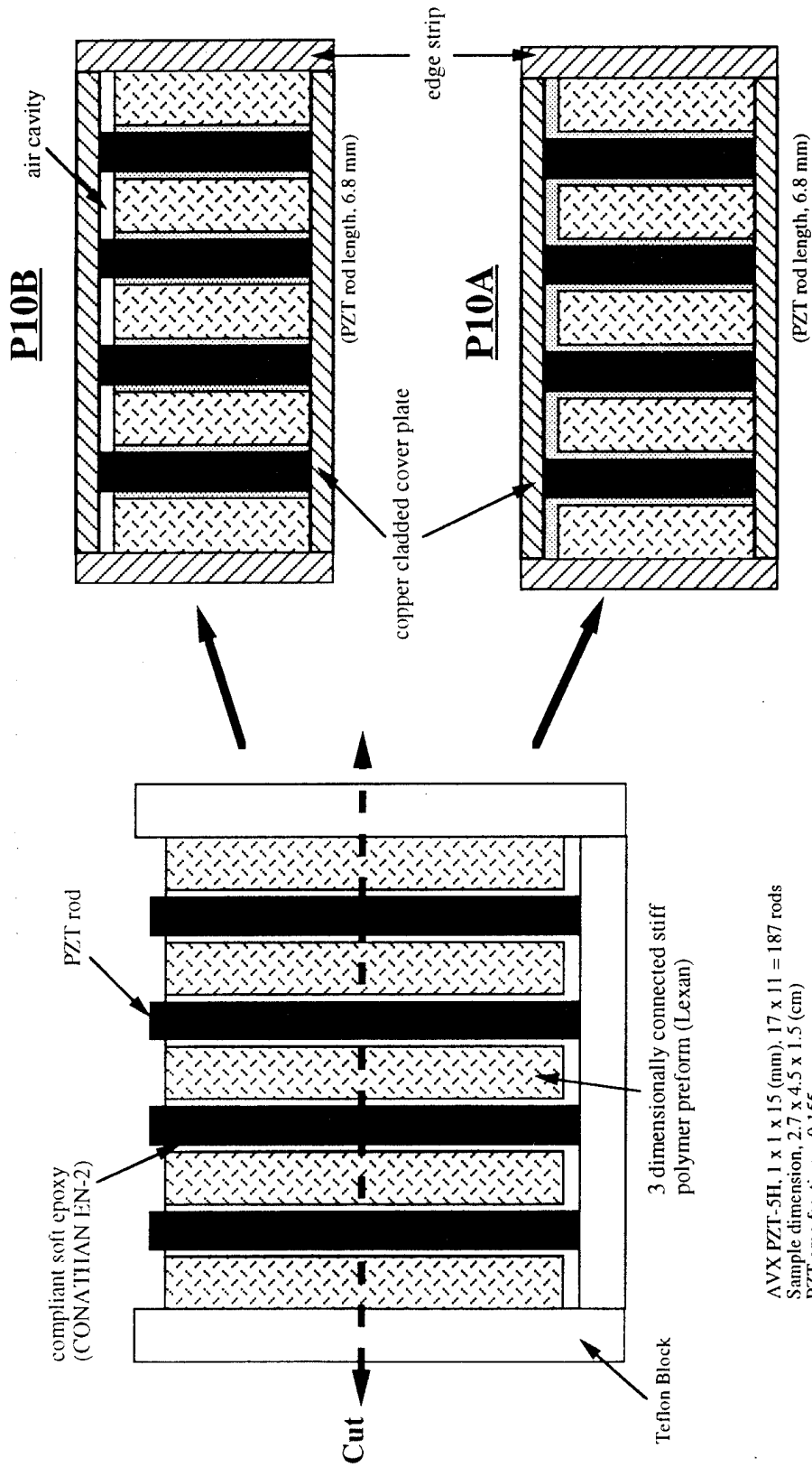


Fig. 9. Schematic of 1-1-3 piezocomposite configuration with (P10B) and without (P10A) the extended PZT rods.

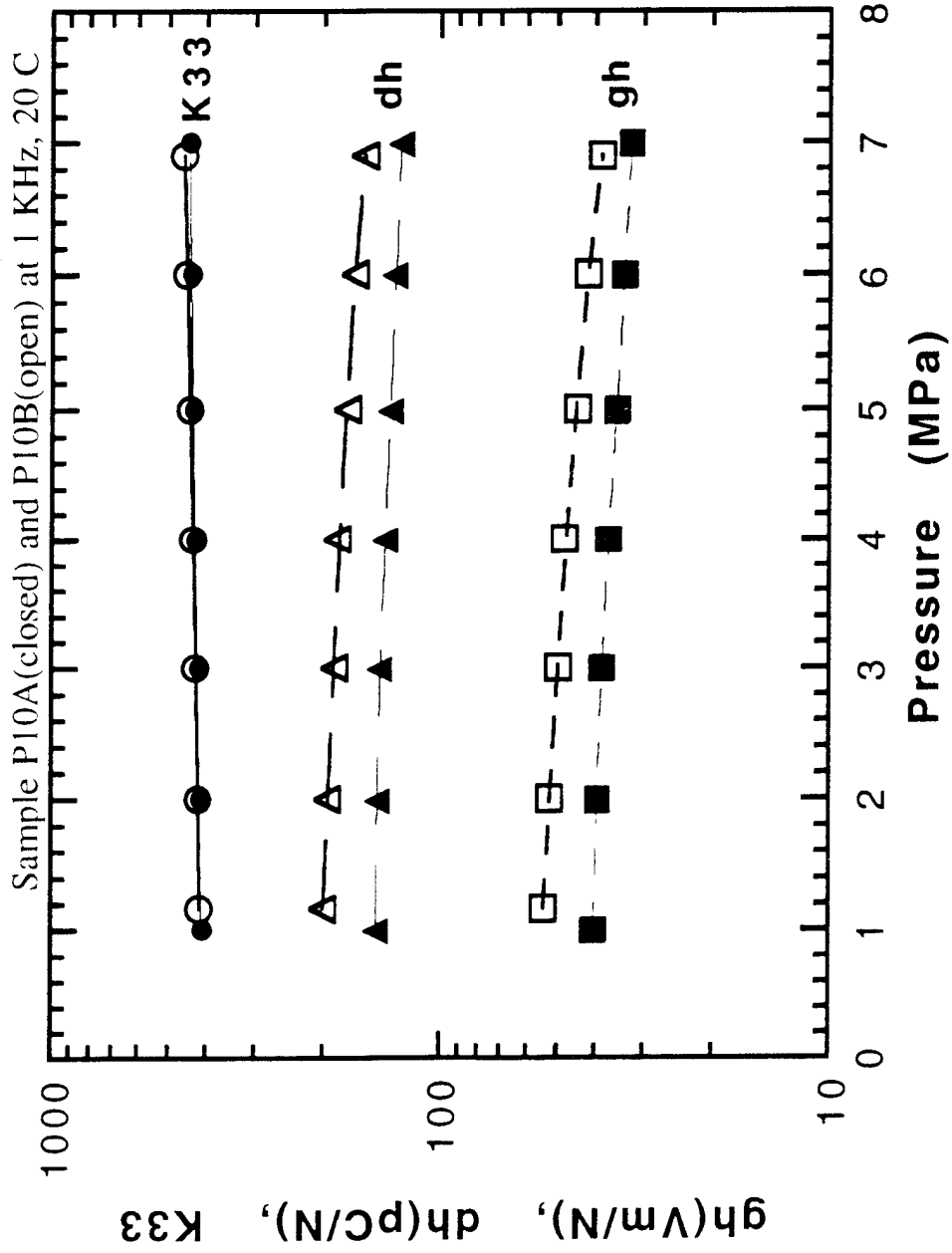


Fig. 10. Pressure dependence of gh, dh and K33. Extended PZT rod composite (P10B) shows enhanced piezoelectric performance.

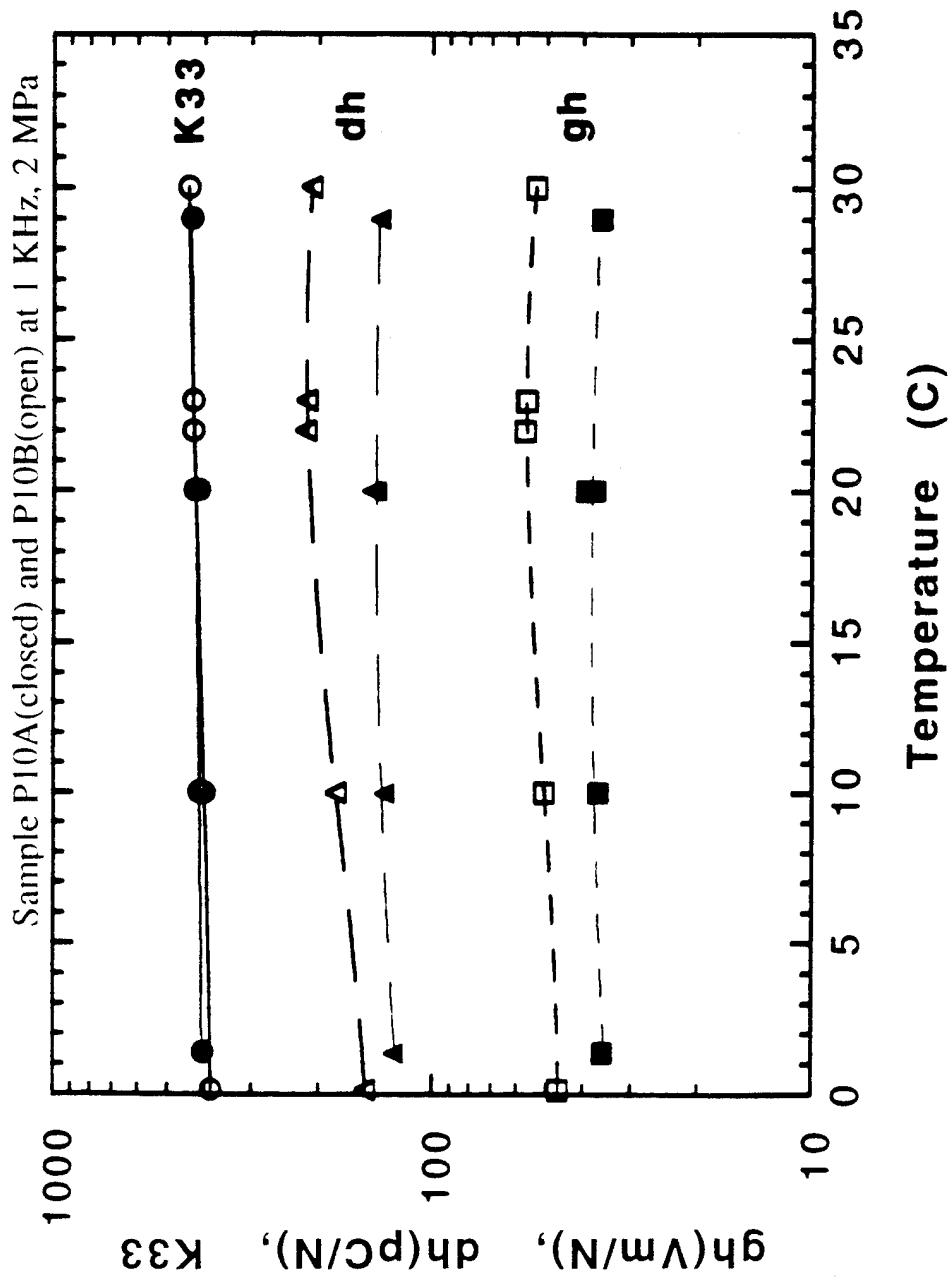


Fig. 11. Temperature dependence of gh, dh and K33. Extended PZT rod composite (P10B) shows enhanced piezoelectric performance.