

AD-A246 662



FMI/JRL-92/001

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DEMONSTRATION OF MANUFACTURING PROCESS  
FOR LARGE AREA, MULTILAYER PIEZOELECTRIC  
COMPOSITES

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QUARTERLY REPORT - JULY 1991 - JANUARY 1992  
CONTRACT NO. N00014-91-C-0091

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February 1992

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92-04242



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SECURITY CLASSIFICATION OF THIS PAGE

REPORT DOCUMENTATION PAGE				Form Approved OMB No. 0704-0188	
1a. REPORT SECURITY CLASSIFICATION Unclassified		1b. RESTRICTIVE MARKINGS			
2a. SECURITY CLASSIFICATION AUTHORITY		3. DISTRIBUTION / AVAILABILITY OF REPORT			
2b. DECLASSIFICATION / DOWNGRADING SCHEDULE					
4. PERFORMING ORGANIZATION REPORT NUMBER(S) FMI/JRL-92/001		5. MONITORING ORGANIZATION REPORT NUMBER(S)			
6a. NAME OF PERFORMING ORGANIZATION Fiber Materials, Inc.		6b. OFFICE SYMBOL (if applicable)	7a. NAME OF MONITORING ORGANIZATION Office of Naval Research		
6c. ADDRESS (City, State, and ZIP Code) 5 Morin Street Biddeford Industrial Park Biddeford, ME 04005		7b. ADDRESS (City, State, and ZIP Code) 800 North Quincy Street Arlington, VA 22217-5000			
8a. NAME OF FUNDING / SPONSORING ORGANIZATION Office of Naval Research		8b. OFFICE SYMBOL (if applicable)	9. PROCUREMENT INSTRUMENT IDENTIFICATION NUMBER		
8c. ADDRESS (City, State, and ZIP Code) 800 North Quincy Street Arlington, VA 22217-5000		10. SOURCE OF FUNDING NUMBERS			
		PROGRAM ELEMENT NO.	PROJECT NO.	TASK NO.	WORK UNIT ACCESSION NO.
11. TITLE (Include Security Classification) Demonstration of Manufacturing Process for Large Area Multilayer Piezoelectric Composites					
12. PERSONAL AUTHOR(S) Lennox, James, Darrah, Shirley, Batha, Dean					
13a. TYPE OF REPORT Quarterly		13b. TIME COVERED FROM 7/91 TO 1/92	14. DATE OF REPORT (Year, Month, Day) 92 Jan 15		15. PAGE COUNT 12
16. SUPPLEMENTARY NOTATION					
17. COSATI CODES			18. SUBJECT TERMS (Continue on reverse if necessary and identify by block number)		
FIELD	GROUP	SUB-GROUP			
19. ABSTRACT (Continue on reverse if necessary and identify by block number)  FMI <sup>®</sup> is conducting a program to demonstrate the capability of economical automated fabrication of fiber reinforced 1-3 piezoelectric composite materials. A series of test specimens is being fabricated and tested to establish relationships between composite structure and performance. The results will be used to design panels in a 30.5 x 30.5 cm (12" x 12") multilayer structure. A modeling study at Stanford University is also being supported by specimen fabrication and testing at FMI. Four specimens from the basic set have been completed and twenty-four more are in production. Eighteen specimens have also been prepared for the Stanford task. Further development has occurred in processing methods including a study of resin systems, development of poling procedures and establishment of quality control standards for PZT rods.					
20. DISTRIBUTION / AVAILABILITY OF ABSTRACT <input type="checkbox"/> UNCLASSIFIED/UNLIMITED <input checked="" type="checkbox"/> SAME AS RPT. <input type="checkbox"/> DTIC USERS			21. ABSTRACT SECURITY CLASSIFICATION Unclassified		
22a. NAME OF RESPONSIBLE INDIVIDUAL J.R. Batha			22b. TELEPHONE (Include Area Code) (207) 282-5911		22c. OFFICE SYMBOL

DD Form 1473, JUN 86

Previous editions are obsolete.

SECURITY CLASSIFICATION OF THIS PAGE

Unclassified

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NWW 2/28/92

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## INTRODUCTION

Fiber Materials, Inc. (FMI<sup>®</sup>) has initiated a program to demonstrate the capability of economical, automated fabrication of fiber reinforced 1-3 piezoelectric composite materials. Ultimately, a multilayer structure containing two panels, each 30.5 x 30.5 cm (12" x 12") will be prepared using the automated techniques which will be used in full-scale production. Prior to fabrication of the multi-layer structure, a series of test specimens, 8.9 cm (3.5") diameter, will also be fabricated to establish relationships between composite structure and performance. The results obtained from tests of the small specimens will be used to design the panels in the multilayer structure. In a separate task of this program a modeling study is being conducted by Prof. B. Auld and Dr. J. Hossack of Stanford University. This is being supported by specimen fabrication and testing at FMI.

The first task involves fabricating several 8.9 cm (3½") diameter PZT composite disks in various architectures so that this material can be tested and characterized. Several variables will be studied including PZT rod size, rod concentration, rod aspect ratio, resin properties, and fiber reinforcement type. These parameters will be varied in the fabrication of 55 test disks. PZT-5H rods sizes of 1.0, .75, and .50 mm and PZT concentrations of 5, 12, and 20 volume % will be used in manufacturing these samples. The aspect ratio will also be varied by changing the thickness of the sample. The epoxy resin systems to be used in fabrication of these disks include HD-68, HD-77 and HD-85J. The mechanical and electrical properties of these resin formulations developed and tested at FMI are listed in Table 1. Two fiber reinforcement types will be employed. S-2 glass roving woven between PZT rod rows in both the X and Y direction will be used on most samples. Random ceramic fibers

TABLE 1

MATRIX RESINS DEVELOPED FOR PZT COMPOSITES

FORMULATION	HD-35	HD-50	HD-68	HD-77	HT-85J	HD-85V	MOD FLEX
<b>MATERIALS</b>							
Epon 815	100	---	---	---	---	---	---
Epon 828	---	100	100	100	100	100	100
RD-2	---	30	40	---	---	15	---
LP-3	---	---	---	---	---	---	100
Versamid 140	198.5	250	225	---	---	40	---
Genamid 2000	---	---	---	---	---	---	35
Jeffamine D-2000	---	---	---	23	---	---	---
Jeffamine T-403	---	---	---	28	40	---	---
Defoamer	Yes	Yes	Yes	Yes	Yes	Yes	Yes
<b>RESIN HANDLING PROPERTIES</b>							
Viscosity, cps @ °C	14500 @ 30	5500 @ 30	6000 @ 34	2200 @ 26	1600 @ 25	4000 @ 32	2200 @ 31
Pot life (150 gm), min	> 180	180	240	> 240	> 240	> 240	27
Peak Exotherm, °C	---	58	52	---	38	---	148
Cure Cycle	Gel @ RT + 1 hr @ 70 °C + 3 hr @ 100 °C	Gel @ RT + 1 hr @ 70 °C + 3 hr @ 100 °C	Gel @ RT + 1 hr @ 70 °C + 3 hr @ 100 °C	Gel @ RT + 2 hr @ 85 °C + 3 hr @ 125 °C	Gel @ RT + 2 hr @ 85 °C + 3 hr @ 125 °C	Gel @ RT + 1 hr @ 70 °C + 3 hr @ 100 °C	Gel @ RT + 3 hr @ 100 °C
ASTM 2240 HARDNESS, TYPE D	32-37	49-53	66-70	77-78	85-86	84-86	43-48
CURED DENSITY @ 25 °C, g/cc	1.043	1.053	1.062	1.141	1.150	1.140	1.213
<b>WATER ABSORPTION @ 25 °C</b>							
After 24 hours, %	9.7	13.6	7.2	0.37	0.17	0.22	0.41
After 1 month, %	24.9	24.6	20.9	1.47	0.74	1.15	2.0
<b>MECHANICAL PROPERTIES @ 25 °C</b>							
Tensile Strength, MPa (psi)	0.490 (71)	0.959 (139)	2.007 (291)	5.000 (725)	47.66 (6910)	35.93 (5210)	3.069 (445)
Tensile Modulus, MPa (psi)	0.986 (143)	2.786 (404)	5.407 (784)	247.59 (35900)	2758.6 (40000)	2572.4 (373000)	24.62 (3570)
Tensile Strain to Failure, %	77	43	50	23	2	2	25
Poisson's Ratio	0.20	0.18	0.25	0.20	0.33	0.33	0.30
<b>ELECTRICAL PROPERTIES @ 25 °C</b>							
Dielectric Constant							
100 Hz	8.46	6.17	5.68	4.57	4.61	4.35	6.81
1 KHz	5.71	5.50	4.77	5.02	4.38	4.35	6.70
10 KHz	4.68	4.75	4.43	4.75	4.17	4.30	6.32
100 KHz	4.16	4.28	4.13	4.54	4.04	4.20	5.96
Dissipation Factor							
100 Hz	0.2	0.25	0.15	0.0	0.05	0.05	0.05
1 KHz	0.17	0.14	0.07	0.03	0.01	0.015	0.04
10 KHz	0.103	0.086	0.051	0.029	0.019	0.015	0.037
100 KHz	0.068	0.065	0.044	0.033	0.026	0.017	0.0463

will be used as reinforcement on 12 of the samples. All samples will be delivered to ONR for testing and characterization.

The test results obtained on the small specimens will be used to design the 30.5 x 30.5 cm (12" x 12") piezelectric composite panels of the multilayer structure. The panels will be separated and surrounded by a layer of dielectric sealant. The entire structure will be surrounded by electrode material.

The second task of this program is to produce 1-3 PZT composites with random and novel patterned arrays of PZT rods parallel to the Z axis, as directed by Professor Auld and Dr. Hossack of Stanford University. Baseline samples will be made with 1 mm PZT rods at a concentration of 10, 20, and 30 volume %. Two different hardness epoxy resin systems will be used and no fiber reinforcement will be employed.

Using the lateral resonance patterns of the impedance curves generated from these parts, the optimum PZT concentration and resin type will be selected for fabrication of random samples. These random samples will include unbalanced rod distributions, random rod placement at different variance levels and matrix variations.

The array patterns generated in this program will affect laterally propagating modes of vibration in a manner quite different from the regular arrays of vertical PZT rods in rows and columns. Designed arrays may give rise to improved directivity characteristic.

This program will investigate the behavior of these random and patterned PZT composites at low frequencies (below 1 MHz). Impedance and permittivity measurements will be provided by FMI. Specimens will be provided to Professor Auld and Dr. Hossack, at their request, for additional measurements.

## RESULTS TO DATE

Most of the starting materials for these composites such as PZT rods, cleaning and treatment chemicals, reinforcement materials, and epoxy resins, have been ordered and received. Four of the 8.9 cm (3½") diameter disks have been completed and impedance spectra have been generated for each sample. For example, the impedance spectrum for P/N 16913 PZT composite disk with 1 mm rods, 5 volume % PZT with HD-68 resin is shown in Figure 1. An additional 24 parts have been started and are at various stages of fabrication. The status of these parts is listed in Table 2.

Originally, urethane resins were to be used for the matrix material of these composite disks. However, calculations performed at Vector Research, Inc. showed that because of high Poisson's ratios they would not perform well as matrix resin in piezoelectric composites. Also, initial test results with the urethane resins indicated that they were too viscous to fully impregnate the glass reinforcement fibers and to degas sufficiently. Also, the pot life of these resins was too short for large scale processing. Therefore, three of the several epoxy resin systems developed by FMI were selected for use in these composites instead. The fourth resin type, if needed, is yet to be determined.

The status of the PZT composites for Stanford University are listed in Table 3. A total of 18 parts have been completed. The six baseline parts were first fabricated, tested and the resulting impedance curves and  $k_t$  values sent to Dr. Hossack for evaluation. Figure 2 shows an example of the strong lateral resonance peak at approximately 300 kHz which Dr. Hossack hopes to attenuate by randomizing rod placement. Four composites were made in which rod locations were unbalanced e.g.: X axis spacing was twice the Y axis spacing, alternate X axis rows were

FIGURE 1

Impedance Spectra Part #16913

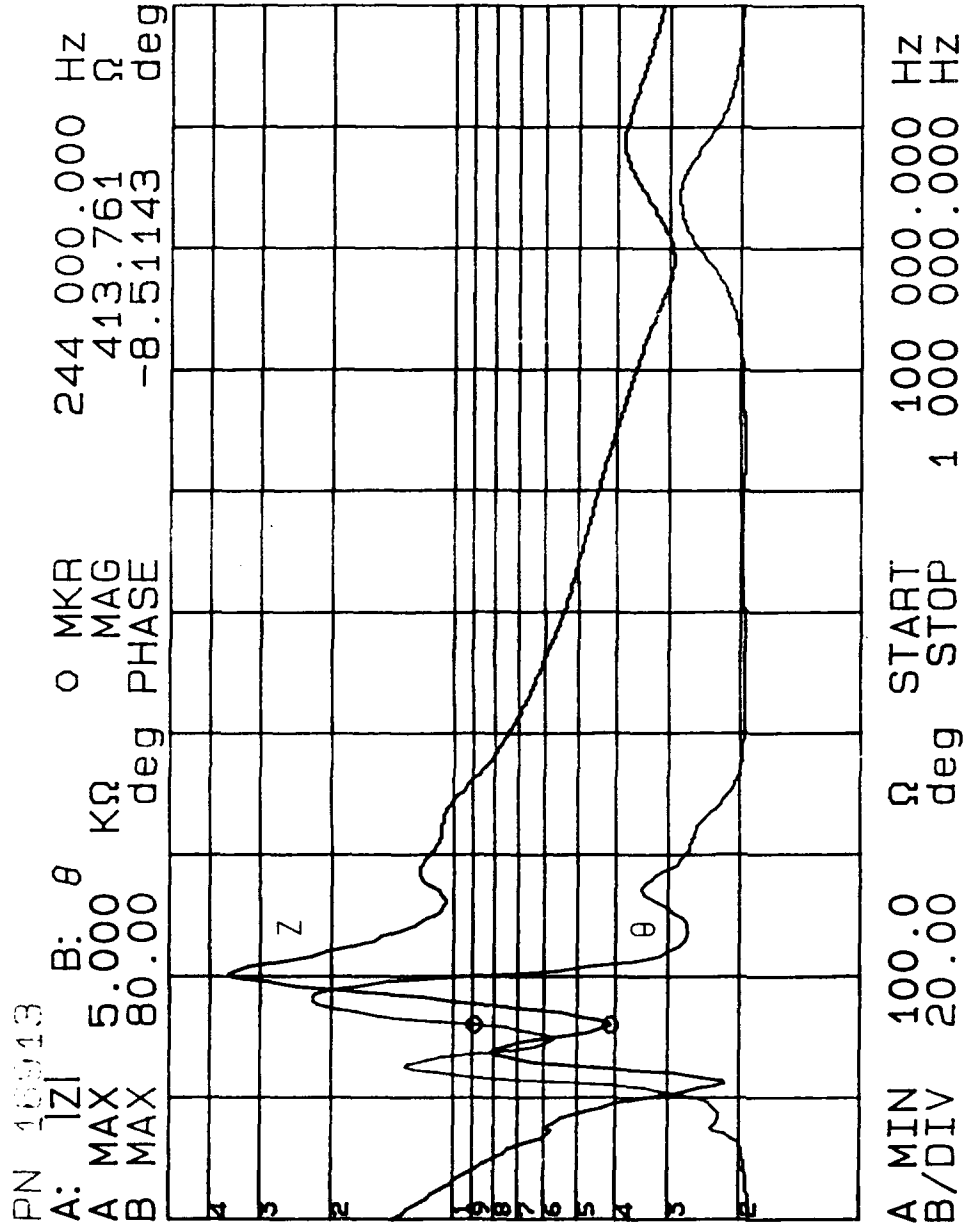


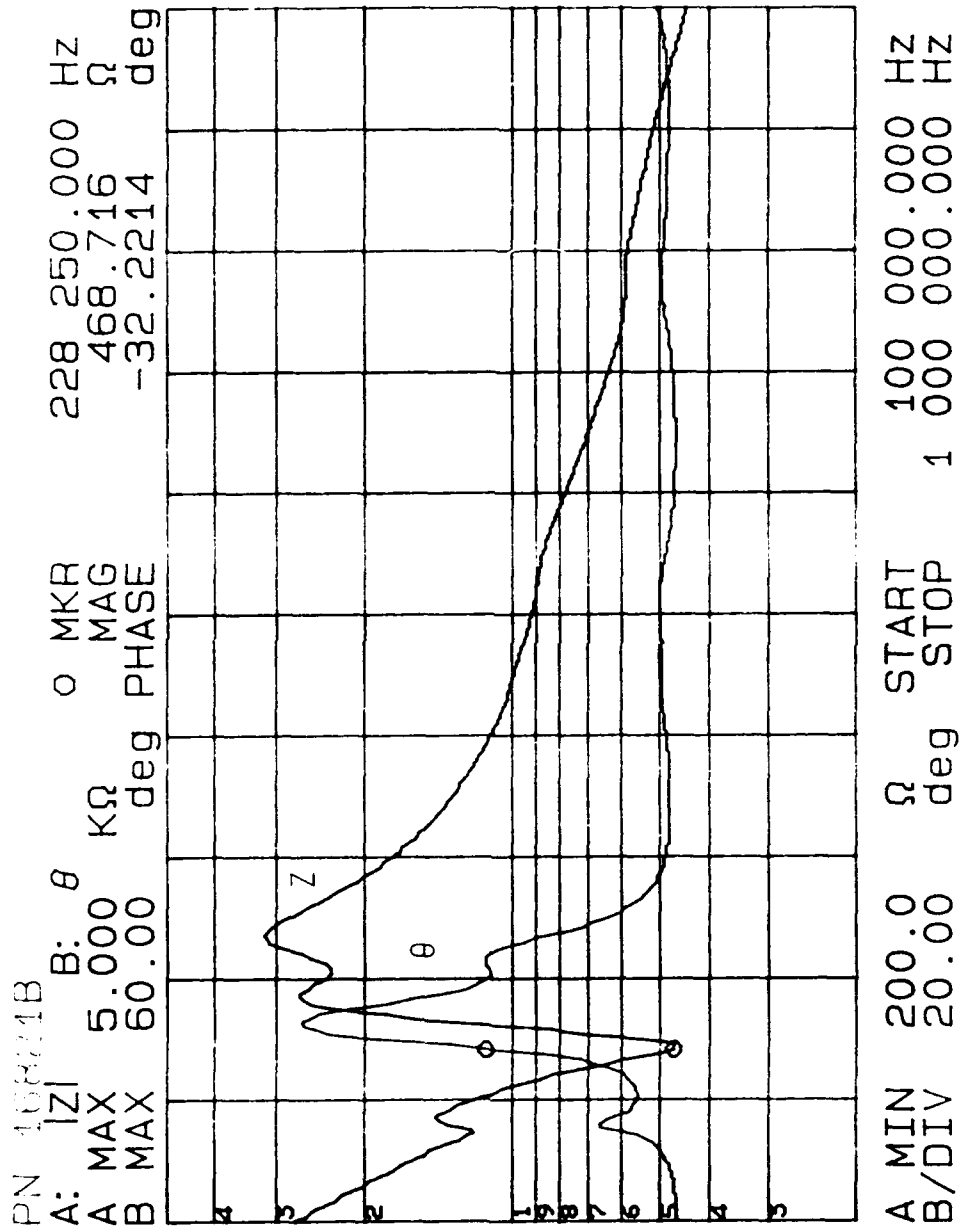
TABLE 2

		ONR PZT COMPOSITE STATUS - DISKS				01-14-92	
PART NUMBER	PART DESCRIPTION	PZT ROD SIZE (mm)	VOL % PZT	RESIN SYSTEM	REINFORCEMENT TYPE	CURRENT STATUS	COMMENTS
16913 A/B	ONR DISK	1.0	10	HD-68	S-2/RANDOM	COMPLETE	
16914 A/B	ONR DISK	1.0	10	HD-77	S-2/RANDOM	MACHINED	
16915 A/B	ONR DISK	1.0	20	HD-85J	S-2/RANDOM	MACHINED	
16916 A/B	ONR DISK	1.0	20	TBD	S-2/RANDOM	WOVEN	
16917 A/B	ONR DISK	1.0	30	HD-68	S-2/RANDOM	COMPLETE	
16918 A/B	ONR DISK	1.0	30	HD-77	S-2/RANDOM	MACHINED	
16919 A/B	ONR DISK	1.0	10	HD-85J	S-2/RANDOM	MACHINED	
16920 A/B	ONR DISK	1.0	10	TBD	S-2/RANDOM	WOVEN	
16921 A/B	ONR DISK	1.0	10	HD-68	S-2/RANDOM	MACHINED	
16922 A/B	ONR DISK	1.0	10	HD-77	S-2/RANDOM	MACHINED	
16923 A/B	ONR DISK	1.0	10	HD-85J	S-2/RANDOM	MACHINED	
16924 A/B	ONR DISK	1.0	10	TBD	S-2/RANDOM	WOVEN	
16925	ONR DISK	1.0	10	HD-68	S-2	IMPREG'D	
16926	ONR DISK	1.0	10	HD-77	S-2	WOVEN	
16927	ONR DISK	1.0	10	HD-85J	S-2	WOVEN	
16928	ONR DISK	1.0	10	TBD	S-2	WOVEN	

TABLE 3

ONR PZT COMPOSITE STATUS - STANFORD PARTS		01-14-92				
PART NUMBER	PART DESCRIPTION	PZT ROD SIZE (mm)	VOL % PZT	RESIN SYSTEM	CURRENT STATUS	COMMENTS
16821	STANFORD BASELINE	1.0	10	HD-50	TESTED	
16822	STANFORD BASELINE	1.0	10	HD-68	TESTED	
16824	STANFORD BASELINE	1.0	20	HD-50	TESTED	
16825	STANFORD BASELINE	1.0	20	HD-68	TESTED	
16826	STANFORD BASELINE	1.0	30	HD-50	TESTED	
16827	STANFORD BASELINE	1.0	30	HD-68	TESTED	
16714	X SPACING TWICE Y	1.0	10	HD-68	TESTED	
16715	ROWS STAGGERED	1.0	10	HD-68	TESTED	
16977	RODS GROUPED IN 4S	1.0	10	HD-68	TESTED	
16978 (1-4)	.020" HOLES IN RESIN	1.0	10	HD-68	TESTED	TO BE ENCAPSULATED
16979 (1-4)	.040" HOLES IN RESIN	1.0	10	HD-68	TESTED	TO BE ENCAPSULATED
16980	STANFORD DESIGN #4	1.0	10	HD-68	TESTED	
16981	RANDOM WITH 10% VAR.	1.0	10	HD-68	DESIGNED	
16982	RANDOM WITH 20% VAR.	1.0	10	HD-68	DESIGNED	
16983	RANDOM WITH 30% VAR.	1.0	10	HD-68	DESIGNED	

FIGURE 2  
 Impedance Spectra Part #16821B



staggered, and rods clustered in groups of four. The samples have been fabricated and test data sent to Stanford University. Two additional samples were fabricated with standard X, Y rod spacing, but holes were drilled in the matrix material between rods in various patterns to reduce lateral resonances. These parts were also completed and tested and will be encapsulated for hydrostatic testing. The next set of samples to be made will consist of composites with PZT rods displaced from their target positions at random distances, based on a Gaussian distribution with 10, 20 and 30% variance. The random locations were generated with a computer program and plotted. These plots have been sent to Dr. Hossack for his approval.

Additional progress has been made in further development of PZT composite processing methods. These improvements include further characterization of epoxy resin systems, optimization of corona discharge poling techniques, and start-up of a quality control procedure for incoming PZT rods.

Work has continued at FMI on developing epoxy resin systems for use in PZT composites. Several resin formulations with various hardness values have been developed that have proven to be suitable for use as a matrix material in these composites. Extensive testing has been done to characterize the mechanical and electrical properties of these resin systems. The results are listed in Table 1.

Corona discharge poling techniques have been developed to replace oil bath poling methods. Advantages of coronal discharge poling include improved flexibility for poling very large parts, reduced incidence of shorting at defect locations within the composite, and the elimination of the possibility of oil contamination of the parts. A corona discharge poling chamber has been set up at FMI and poling procedures optimized such that

PZT rods within a composite can be poled to  $d_{33}$  values at least as high as those resulting from oil bath poling.

Quality control methods are being developed to ensure that incoming PZT rods have acceptable physical and electrical properties. Samplings of incoming PZT rods have been poled and their density, dielectric constant,  $d_{33}$ ,  $k_{33}$ , and  $N_t$  values measured. This information is being used to establish baseline data of PZT rod properties. This data will be used as a guide for evaluating different PZT rod vendors and to check the quality of future PZT rod shipments.

#### FUTURE WORK

Fabrication of the 8.9 cm ( $3\frac{1}{2}$ " ) diameter disks will continue and these composites will be shipped as they are completed. Selection of the fourth resin system will be made per ONR approval so that these parts can continue to be processed. Also, the design of the 30.5 x 30.5 cm (12" x 12") multilayer composite, schedule for fabrication later in the program, will be addressed.

FMI will continue to provide experimental data to Dr. Hossack for use in the design of the next set of composites. The composites designed to have randomly placed PZT rods per a Gaussian distribution at 10, 20 and 30% variance will be fabricated next. Additional samples will be fabricated as directed by Dr. Hossack.

#### SUMMARY

The task of producing  $3\frac{1}{2}$ " diameter PZT composite disks in various architectures for characterization studies is well underway. Four disks have been completed and an additional 24 disks are at various stages of processing. Also, significant

progress has been made in fabricating PZT composite test specimens with random and unique rod placement designs for evaluation at Stanford University. FMI has built and tested 18 of the 40 composite samples required. FMI will continue to fabricate samples per Dr. J. Hossack's designs in an attempt to improve the piezoelectric properties of these composites.

Progress has also been made in the development of PZT composite processing methods. Epoxy resin systems for these composites have been further characterized, corona discharge poling techniques have been optimized, and quality testing procedures for incoming PZT rods have been established. FMI will continue to work to improve fabrication techniques as the required composite disks are processed and tested.

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