01 Jan 1994

Fabrication of 2-2 Connectivity PZT/Thermoplastic Composites for High Frequency Linear Arrays

Wayne Huebner
Missouri University of Science and Technology, huebner@mst.edu

Mary R. Reidmeyer
Missouri University of Science and Technology, maryrr@mst.edu

L. Busse

Jeffry W. Stevenson

Recommended Citation

W. Huebner et al., "Fabrication of 2-2 Connectivity PZT/Thermoplastic Composites for High Frequency Linear Arrays," *Proceedings of the 9th IEEE International Symposium on Applications of Ferroelectrics, 1994*, Institute of Electrical and Electronics Engineers (IEEE), Jan 1994.
The definitive version is available at https://doi.org/10.1109/ISAF.1994.522339

Fabrication of 2-2 Connectivity PZT / Thermoplastic Composites for High Frequency Linear Arrays

W. Huebner, M. R. Reidmeyer, and J. W. Stevenson

Department of Ceramic Engineering
University of Missouri-Rolla
Rolla, Missouri 65401

L. Busse

Tetrad Corporation
Englewood, CO 80112

ABSTRACT

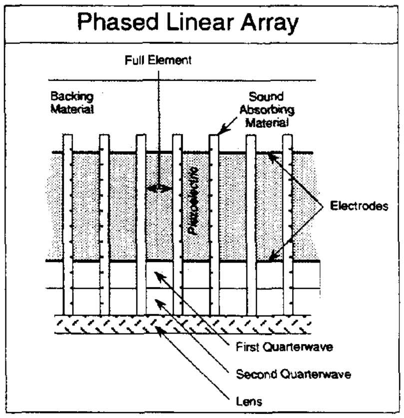
An alternate approach for fabricating PZT/polymer composites with 2-2 connectivity with fine scales is described. Thin (≤20μm) sintered PZT plates and sheets of a thermoplastic polymer film (≤10μm) were bonded together via thermal processing. Stack sintering of tape cast PZT generated the necessary PZT plates, while tape cast polymers were used to control the thermoplastic thickness. Composite blocks were cut to required dimensions for linear arrays, electroded, and poled. Electromechanical properties were measured to evaluate the composites. The significance of this fabrication technique is that it is able to generate 2-2 structures at a scale level unachievable by conventional dice-and-fill fabrication methods.

INTRODUCTION

Piezoelectric composites are now widely used for many ultrasonic transducer applications. Reviews by Guraraja et. al [1], Smith [2-5] and Oakley [6] clearly illustrate the influence of scale, connectivity and symmetry on the properties of composite piezoelectrics, and lend guidance to the transducer engineer for their application. In essence, the biggest advantage of using a piezoelectric composite compared to its monolithic counterpart is its higher electromechanical coupling coefficient, which in turn leads to higher sensitivities and broader bandwidths.

An ongoing trend in medical ultrasonics is to increase the frequency of imaging systems (≥10 MHz), for applications such as phased linear array transducers used for laproscopy and, in the future, intravascular imaging. However, these transducers must have a very fine spacing of the piezoelectric elements (≤50 μm) in order to minimize acoustic clutter associated with grating lobes. In addition, subdivision of the piezoelectric into smaller elements would minimize coupling to unwanted lateral vibrational modes, with concurrent improvements in the thickness coupling coefficient. Currently, the synthesis techniques needed to achieve the requisite scale and periodicity of the composite have become the effective limitation for their exploitation at higher frequencies.

For instance, Figure 1 shows a typical process for fabricating a phased linear array, a modified "dice-and-fill technique" [7]. The finest interelement spacing is controlled by the kerf width of the saw blade, which is currently ≈25-40 μm. The $Pb(Zr,Ti)O_3$ (PZT) can be reliably diced into structures ≈50 μm in size. This scale is sufficient for 10 MHz transducers, but higher frequencies will dictate a further reduction. Considerations of dicing technologies coupled with grain size and strength limitations of the ceramic dictate that new fabrication technologies be developed, which is the subject of this paper.



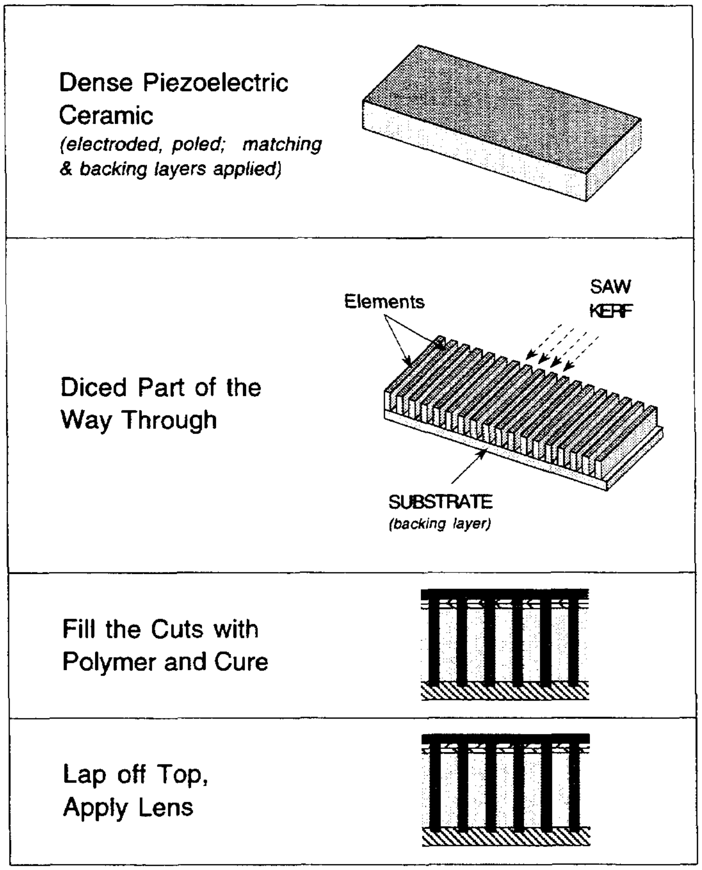


Figure 1. Schematic illustration of the dice and fill technique used to fabricate phased linear arrays.

CH3416-5 0-7803-1847-1/95/$4.00©1995IEEE

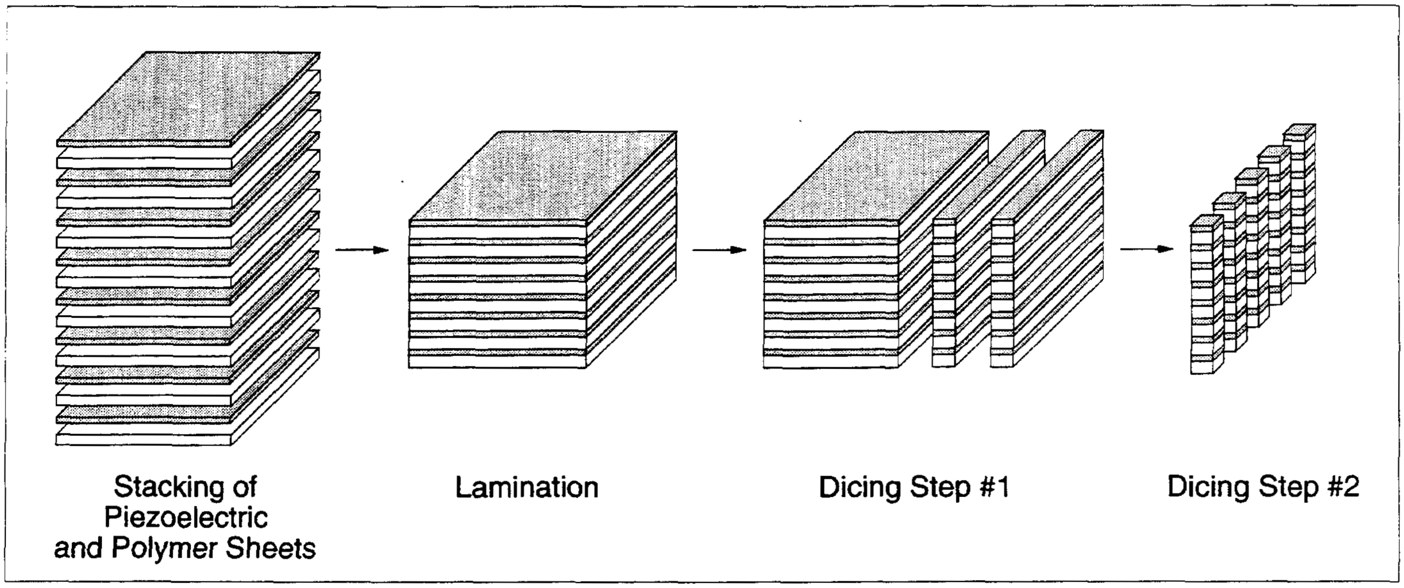


Figure 2. Overview of the process used to synthesize the 2-2 composites.

The process is simple; pre-sintered PZT plates are joined together using a thermoplastic adhesive polymer film to yield a composite with 2-2 connectivity. Dicing is only used to achieve the final transducer geometry. This technique allows for the fabrication of finer scale composites, and is described below.

EXPERIMENTAL PROCEDURE

Sintering of tape cast PZT was used to produce the piezoelectric elements. Slurries were prepared by dispersing a pre-calcined soft PZT powder in a solution containing 17 ml ethanol, 8 ml toluene, 0.5 g Menhaden fish oil, and 3 g polyvinyl butyral. The slurries were de-aired, cast on glass substrates with a single knife doctor blade, and dried at room temperature in air. The PZT tapes were cast at various thicknesses ranging from 30 to 250 μm to obtain sintered thicknesses ranging from 10 to 55 μm. Square sections (2 cm x 2 cm) of the tapes were stack-sintered using polished PZT setters (85 μm thickness), $PbZrO_3$ powder as the lead source, and covered with an Al_2O_3 crucible. Binders were burned out in air at 450°C, followed by sintering at 1250°C for 30 minutes. The PZT setters maintain a high PbO activity around the tapes and result in flat elements.

Polyvinyl formal (PVF) was chosen as the thermoplastic due to its excellent adhesive strength to PZT. A PVF solution was prepared for tape casting by dissolving it in ethanol and toluene. After being cast on glass substrates with the doctor blade at varying thicknesses, PVF films were dried at room temperature in air. The thickness of the films was adjusted between 9 and 20 μm by varying the casting thickness.

Figure 2 exhibits the procedure for preparing the composites. Sintered PZT plates and PVF films were alternately stacked and then laminated by heating (210°C) the stacks in a vacuum oven under uniaxial compression (0.1MPa). Transducers were sliced out of the composite block using a dicing saw (Kulicke & Soffa Industries, Model 775 Wafer Saw), electroded with sputtered gold, then poled at room temperature at 25 kV/cm for 30 minutes. Resonance and dielectric measurements were performed with an impedance analyzer (Hewlett-Packard 4194A Impedance Analyzer with a 41941A Impedance Probe and 16092A Test Fixture).

RESULTS AND DISCUSSION

This fabrication technique dictates that the sintered PZT plates are thin and flat, and that the thermoplastic polymer has good adhesion to the PZT. Using stack sintering, PZT plates with thicknesses between 10 and 55 μm were easily achieved, in all instances for substrates 1.5 x 1.5 cm. Larger area plates were difficult to handle, but considerations of the final transducer size make it unnecessary to make larger composite blocks. Densities were ≥98% theoretical, and grain sizes were ≈3-5 μm. During initial studies the piezoelectric properties of individual plates were evaluated to make sure the sintering process did not result in inactive surfaces. Poling was easily achieved without breakdown, with d_{33} coefficients ≥500 pC/N, and a $k_p \approx 60\%$ (thickness mode resonance too high to measure). Hence this process allows for the synthesis of thin PZT elements as well as independent control over the grain size. Other strategies for fabricating structures at this scale level also result in smaller grain sizes, which is detrimental to the piezoelectric properties of PZT ceramics [8].

Figure 3 shows a series of SEM micrographs of lapped composites with differing PZT and PVF thicknesses. Clearly the PVF provided excellent adhesion to the PZT plates; bonding at the PZT/polymer interfaces was uniform with only a few noticeable voids or delaminations. The PZT elements were not fractured. Note the smallest thickness of the PVF is ≈9 μm; this results in an interelement spacing that is three times closer than can be achieved using dicing.

The electrical performance of the composites of Figure 3 are contained in Table I. Values of the thickness electromechanical coupling coefficients of the composites, $\overline{k_t}$, were calculated from the impedance measurements using [9]:

$$\overline{k_t}^2 = \frac{\pi}{2}\frac{f_{min}}{f_{max}}\tan\left(\frac{\pi}{2}\frac{f_{max}-f_{min}}{f_{max}}\right) \qquad (1)$$

where f_{min} and f_{max} are the frequencies of minimum and maximum impedance corresponding to the thickness resonant mode. The mechanical quality factor, Q_m, was calculated using [9]:

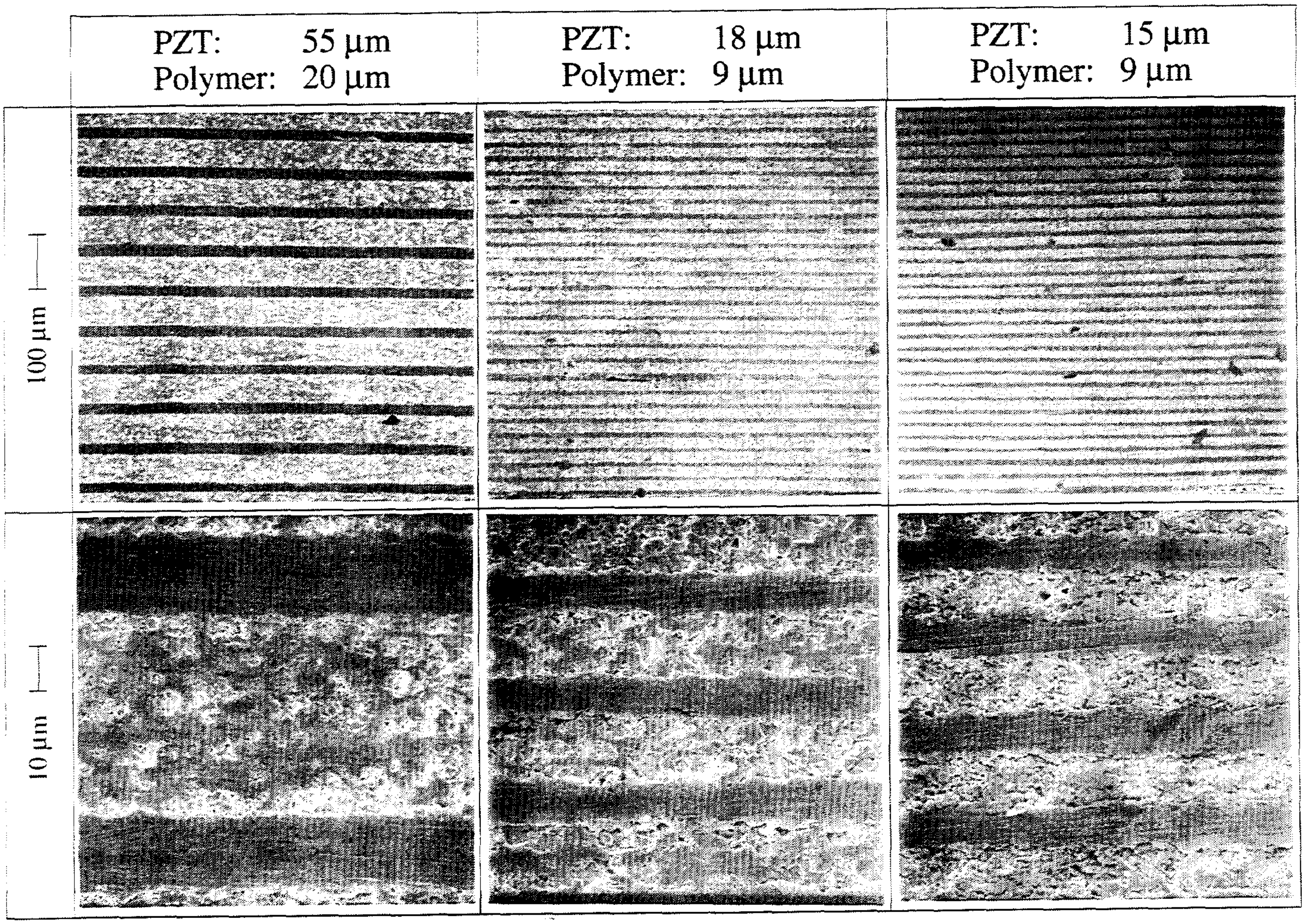


Figure 3. SEM micrographs of several PZT/PVF composites with varying PZT and PVF thicknesses.

$$Q_m = [4\pi \cdot \Delta f \cdot |Z_m| \cdot C]^{-1} \qquad (2)$$

where $\Delta f = f_{max} - f_{min}$, $|Z_m|$ is the magnitude of the electrical impedance at f_{min}, and C is the capacitance measured at 1 kHz.

Figure 4 exhibits the thickness mode impedance and phase angle plot, for the third composite shown in Figure 3. This composite was 250 µm thick, with 105 elements composed of 15 µm thick PZT elements separated by 9 µm thick layers of PVF. The thickness mode resonance is sharp, with $\overline{k_t}=0.68$. The lower frequency harmonics are due to the planar mode resonance of the PZT bars ($\approx$ 400 kHz). For an actual transducer the overall dimensions would move the planar resonance closer to the thickness mode, and hence could be a problem. Materials with a high k_t/k_p ratio such as a modified lead titanate compositions would minimize this problem, albeit at the expense of sensitivity.

As shown in Table I, values of $\overline{k_t}$ for the composites ranged between 0.63-0.68. These high values of $\overline{k_t}$, are similar to values of k_t reported for PZT/epoxy composites prepared by the dice-and-fill technique [10] and approach the k_{33} value for soft PZT compositions. The high electromechanical coupling shown by these composites make them excellent candidates for linear array transducer applications operating at frequencies ≥ 10 MHz.

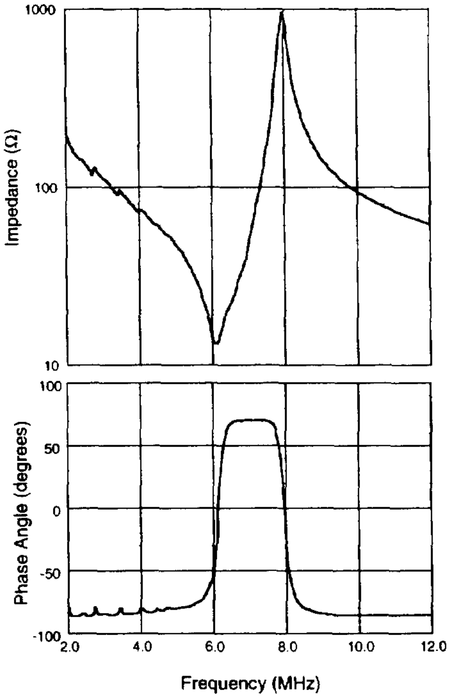


Figure 4. Thickness mode resonance results for a 250 µm thick, composite with 105 elements composed of 15 µm thick PZT elements separated by 9 µm thick layers of PVF.

TABLE I

ELECTROMECHANICAL PROPERTIES OF PZT/PVF COMPOSITES

# of PZT Elements	PZT thickness (μm)	PVF thickness (μm)	k_t	Q_m
60	55	20	0.63	5.5
105	15	9	0.64	5.1
105	18	9	0.68	5.1

CONCLUSIONS

A simple technique for fabricating PZT/polymer composites was developed which allows for the fabrication of 2-2 composites on a finer scale than allowed by dice-and-fill techniques. PZT plates with sintered thicknesses as low as 10 μm were produced. The thickness of the polymer could be reduced down to 9 μm, which yields an interelement spacing three times smaller than that attainable with the dice-and-fill technique. This technique offers the additional advantage of being able to build aperiodic structures by simply changing the thickness of the polymer film. By adding filler materials to the polymer, one would also expect the attenuation of the intervening polymer could be increased, which would decrease crosstalk between elements.

Acknowledgments

Financial support was provided by Tetrad Corporation and N.I.H. Grant HL 44230.

REFERENCES

[1] T.R. Gururaja, A. Safari, R.E. Newnham and L.E. Cross, "Piezoelectric Ceramic-Polymer Composites for Transducer Applications," pp. 93-128 in Electronic Ceramics, ed. L.M. Levinson, Marcel Dekker, New York, New York (1987).

[2] W.A. Smith and A.A. Shaulov, "Composite Piezoelectrics: Basic Research to a Practical Device," *Ferroelectrics*, **87**, 309-320 (1988).

[3] W.A. Smith, "The Role of Piezocomposites in Ultrasonic Transducers," p. 755-766, *Proceedings of the 1989 IEEE Ultrasonics Symposium* (1989).

[4] W.A. Smith, "The Application of 1-3 Piezocomposites in Acoustic Transducers," pp. 145-152, *Proceedings of the 1990 IEEE Symp. on the Applications of Ferroelectrics*, (1991).

[5] W.A. Smith, "The Key Design Principle for Piezoelectric Ceramic / Polymer Composites," pp. 825-838, Recent Advances in Adaptive and Sensory Materials and Their Applications, ed. by C.A. Rogers and R.C. Rogers, Technomic Publishing, Lancaster, PA (1992)

[6] C.G. Oakley, "Analysis and Development of Piezoelectric Composites for Medical Ultrasound Transducer Applications," Ph.D dissertation, Penn State University (1991).

[7] H.P. Savakus, K.A. Klicker, and R.E. Newnham, "PZT-Epoxy Piezoelectric Transducers: A Simplified Fabrication Procedure," Mat. Res. Bull., 16, 677-680, 1981.

[8] A.H. Webster and T.B. Weston, "The Grain-Size Dependence of the Electromechanical Properties in Lead Zirconate-Titanate Ceramics," J. Can. Cer. Soc., 37, 41-44, 1968.

[9] IRE Standards on Piezoelectric Crystals: Measurements of Piezoelectric Ceramics, 1961.

[10] H. Takeuchi and C. Nakaya, "PZT/Polymer Composites for Medical Ultrasonic Probes," Ferroelectrics, 68, 53-61, 1986.