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Highly Anisotropic Piezoceramics

Mike Staines
Robinson Research Institute
Victoria University of Welington
Lower Hutt, New Zealand
Mike.Staines@vuw.ac.nz

Jasmine Kennedy
Robinson Research Institute
Victoria University of Welington
Lower Hutt, New Zealand
Jasmine.Kennedy@vuw.ac.nz

Gideon Gouws
School of Engineering
Victoria University of Welington
Wellington, New Zealand
Gideon.Gouws@vuw.ac.nz

Frederic LeCarpentier
Callaghan Innovation
Lower Hutt, New Zealand
Frederic.Lecarpentier@
callaghaninnovation.govt.nz

Eugene Stytsenko
Callaghan Innovation
Lower Hutt, New Zealand
Eugene.Stytsenko@
callaghaninnovation.govt.nz

Paul Harris
Callaghan Innovation
Lower Hutt, New Zealand
Paul.Harris@
callaghaninnovation.govt.nz

***Abstract*—This work describes the fabrication process to realise a highly columnar porous microstructured piezoceramic, and provides 2D array element pulse response plots for both porous and nonporous material.**

Keywords—microstructure, piezoceramic, anisotropy

I. Introduction

2D transducer arrays are usually produced with the dice and fill technique, with sub-dicing used to limit unwanted element modes; dice and fill is a major cost in device manufacture. A piezoelectric material that features a well-developed columnar microstructure would circumvent this by implicitly providing superior modal characteristics, and reduce the acoustic impedance for efficient coupling. Ultimately, with a high microstructure density, fabrication by electrode patterning alone may be adequate and provide very low-cost manufacture.

Some piezoelectric materials (e.g. lead metaniobate) exhibit porosity but as the pores are somewhat randomly oriented wave propagation is largely isotropic. Material with anisotropic porosity has previously received some attention because of the potential for enhanced properties. Examples include the assembly of columnar structures of $PbTiO_3$ particles in an epoxy matrix using dielectrophoresis [1], effectively reproducing on a microstructural scale the 1-3 composite connectivity of a dice and fill pillar array. Freeze texturing [2] has been used to produce a lamellar microstructure in PZT. Oriented pore microstructure has been produced in alumina ceramics by aligning polymer fibres in the green ceramic body and burning out the fibres during sintering. Isobe et al [3] aligned carbon fibres by co-extrusion with a plastic ceramic paste. Miyagawa and Shinohara [4] used a magnetic field to align nickel wires and nickel-coated carbon fibres in a ceramic slip.

Here we describe the steps taken to realise an anisotropic piezoceramic material characterized by a porous columnar microstructure at the micron scale.

II. Material Preparation Method

Our oriented pore piezoceramic material was produced using a process based on the method described by Miyagawa and Shinohara [4].

Fig. 1 shows the sequence of process steps:

1. Nickel coated fibres are dispersed in an aqueous suspension of PZT powder
2. The fibres rotate to align parallel to the magnetic field
3. The field gradient drags the fibres towards the high field region at the substrate
4. A dense array of fibres assembles on a porous substrate
5. The ceramic slip deposits on the substrate, with consolidation assisted using a vacuum applied to the underside of the substrate
6. The consolidated ceramic is dried, then fired, burning out the polymer fibres to leave an oriented pore array.

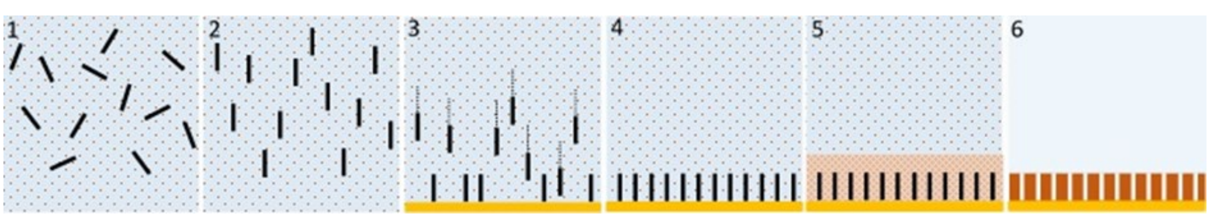

Fig. 1 Process for producing oriented pore ceramic material by magnetic alignment of nickel-coated fibres as described in the text

We used 7 and 5 µm diameter carbon fibre, with lengths 1.0 and 0.7 mm, with nickel coating around 0.1 µm thick, supplied by Conductive Composites Inc. The mold was positioned in the gap of a magnet consisting of an iron yoke and a cylindrical NdFeB permanent magnet. The magnetic field at the substrate was around 0.4 T, with the field gradient above the substrate around 10 T/m.

Alignment of the fibres was firstly demonstrated using fibres dispersed in low-viscosity epoxy (Epotek 301) which has the advantage, compared to the ceramic dispersion, that the fibre motion and its distribution can be observed optically during the process.

The ceramic powder was type 855 PZT supplied by APC, the maximum grain size of the powder is about 2 µm. Before use we removed the binder from the powder with a 650°C heat treatment in flowing air. The resulting green PZT discs had diameter 24 mm and thickness 4 mm. After drying the green ceramic disc is separated from the substrate and undergoes heat treatment; binder burnout at 600°C followed by sintering in air at 1250°C. Thermogravimetric measurements show that the burnout of the fibres proceeds in two stages in air; oxidation of the nickel coating at 400°C to 550°C, followed by oxidation of the carbon fibre at 600°C to 840°C. As supplied, the APC 855 powder has significant nickel content, so much so that the minor addition from the nickel coating of the fibres has no apparent impact on sintering the PZT. The sintered disc is then polished to 2 mm thickness for scanning electron microscope (SEM) inspection. To preserve the pore array only the minimum of material is removed when polishing the bottom (pore) surface.

III. Ultrasonic Characterisation Method

In establishing the ultrasonic performance of the anisotropic piezoceramic material it was decided not to use a conventional transducers approach, which for instance has backing materials which would be frequency sensitive and influence the pulse response, but rather use the material alone simply bonded to an aluminium bronze acoustic delay line 8.5 mm thick. Aluminium bronze was selected due to its ready availability and because its acoustic impedance is close to that of the oriented pore PZT.

The material was fabricated into a 2D array transducer for ultrasonic characterisation. The 24 mm diameter PZT disc was coated with silver paste (Gwent Electronic Materials Ltd, No 2000107P3), which was fired on by heating in a controlled ramp to 750 °C. After 15 min at 750°C the disk was cooled down and placed into an oil bath at 110 °C. The sample was then poled to induce a piezoelectric response by applying a 2.5kV/mm electric field across its thickness for 30 min. The 2 mm thick disc was machined to be flat and parallel, and then the porous face diced (kerf 30 µm, pitch 750 µm, depth of kerf 700 µm) and infiltrated with a 2 µm silica bead epoxy composite, then machined flat again. This diced face was metalized using e-beam evaporation (30nm titanium, 300nm silver) and bonded to the aluminium bronze. The piezoceramic was then lapped to a thickness of 570 µm, corresponding to a design frequency of 4.3 MHz to 5 MHz i.e. similar to that used in common medical ultrasonic imaging. The exposed face was then metalized (e-beam evaporation as before).

Transducers were fabricated from wafers of three different type 855 materials, (1) conventional isotropic material supplied to us by APC, and then wafers produced by our slip casting process with (2) a very low pore density of around 50 mm^{-2}, and (3) with a pore density of around 1000 mm^{-2}.

Two transducers were fabricated using the APC supplied wafers. The first was diced at 750 µm pitch as described above. Because of the somewhat cubic nature of the elements it would be expected to exhibit poor performance due to modes within the element. In normal practice the designer would sub-dice each element. Accordingly, the second isotropic disc was sub-diced by a factor of four (x and y).

As shown in Fig. 4, an industry-standard Olympus 5073PR pulser-receiver is used to both stimulate the material and to detect the ultrasonic signal emitted from the material, we use an Olympus V543 device as detector. Contact with the elements uses a 30 µm copper foil that has been adhered to a highly porous rigid foam, electrical contacts can be reliably made whilst retaining an ‘air backed’ design i.e. the oriented pore material properties alone are exhibited in the test without being aided by a backing. A Yokogawa oscilloscope is used to capture the data.

IV. Results and Discussion

Fig. 2 shows images of fibre alignment in epoxy. Excellent alignment was attained with only a few fibres retained on the container wall (none in the bulk).

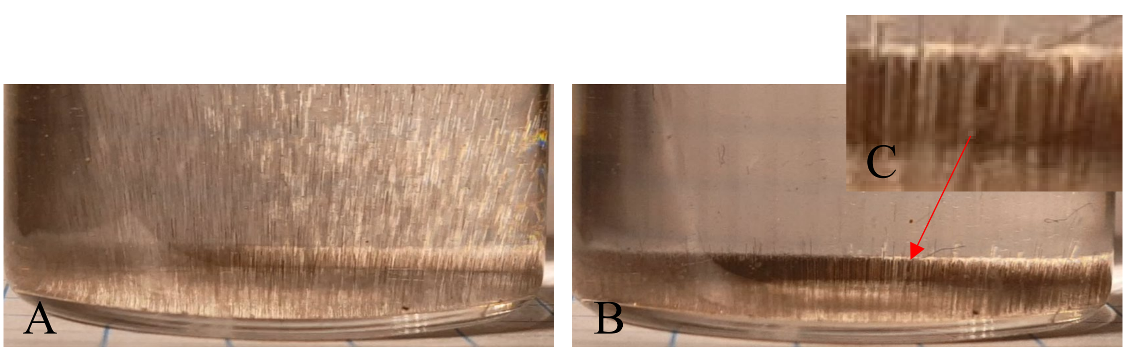

Fig. 2. Fibres in PZT slurry mimicking solution, A. 2 sec post filling container, B. at 20 sec, C. magnified image of area indicated.

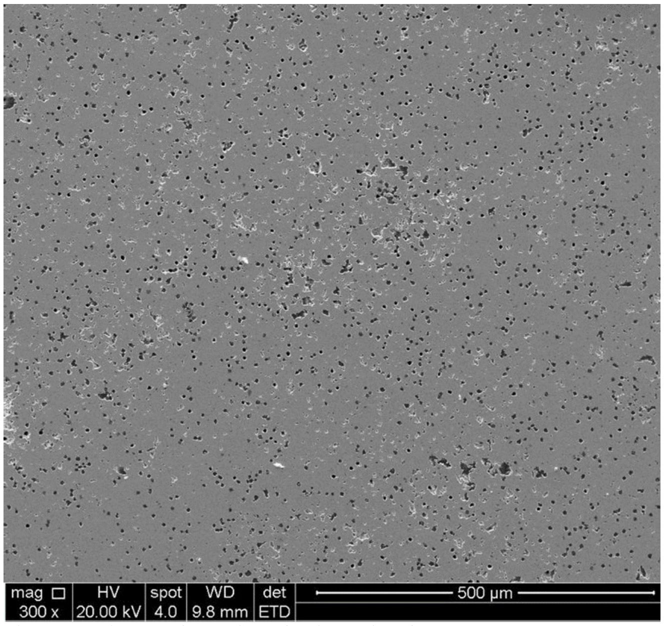

Fig. 3 SEM image of a section of a sintered wafer

The SEM image, Fig. 3, shows the round pores left by burning out the fibres are easily distinguished from a background of more isotropic voids. The pore density for this sample is 1050 mm^{-2}. Pore densities as high as 2000 mm^{-2} have been obtained in some samples.

Fig. 4 shows the ultrasonic measurement setup and Figs. 5 - 8, are screen plots of the acoustic signatures measured for transducers fabricated from different wafers (horizontal 500 nsec/div, vertical 100 mV/div, 20 MHz bandwidth, no average).

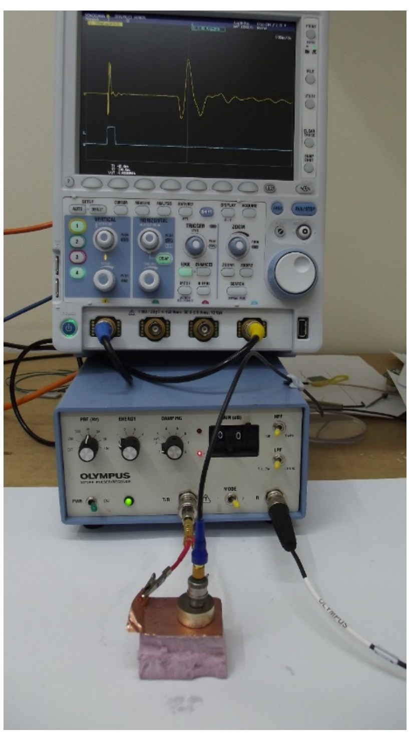

Fig. 4 Ultrasonic measurement setup

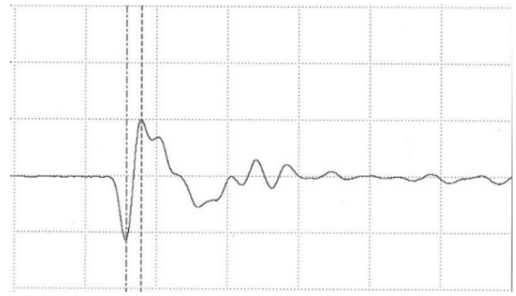

Fig. 5 Pulse response (500 nsec/div, 100 mV/div) of manufacturer-supplied APC type 855 piezoelectric wafer diced at 750 µm pitch. This material was prepared by a standard pressing and sintering process.

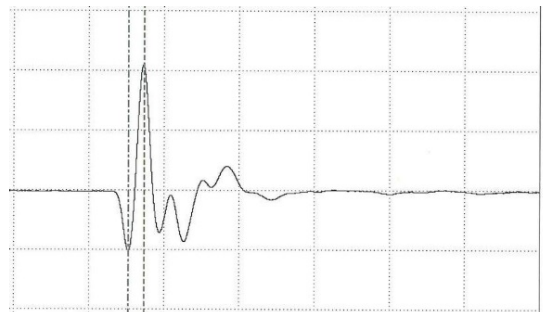

Fig. 6 Pulse response (500 nsec/div, 100 mV/div) of manufacturer-supplied APC type 855 piezoelectric wafer diced at 750 µm pitch, with factor of 4 sub-dicing. Note the much cleaner response following the leading edge of the pulse compared to Fig. 5 without sub-dicing.

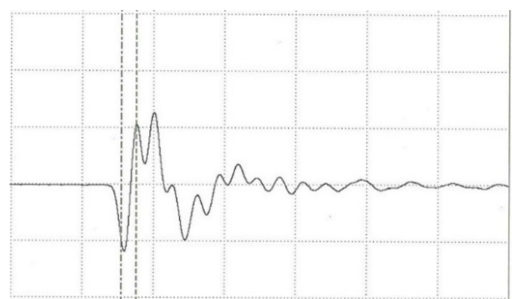

Fig. 7 Pulse response (500 nsec/div, 100 mV/div) of type 855 piezoelectric wafer prepared by using the vacuum-assisted slip casting procedure with a low pore density 50 mm^{-2}, diced at 750 µm pitch.

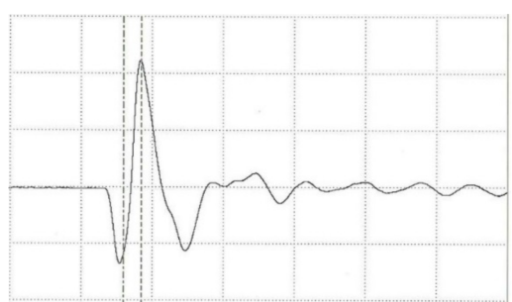

Fig. 8 Pulse response (500 nsec/div, 100 mV/div) of type 855 piezoelectric wafer prepared using the vacuum assisted slip casting process with pore density 1050 mm^{-2}, diced at 750 µm pitch.

The first plot, Fig. 5, shows the performance of the manufacturer-supplied disc of the 855 material, i.e. without oriented pores. The wave shape exhibits modes, revealed by the low frequency components in the waveform occurring after the cursors positioned at the leading pulse edges. The operating frequency is 4.3 MHz.

It would be normal practice for the transducer designer to use sub-dicing of the element to limit the element low frequency mode amplitude, and this is shown in Fig. 6. It should be noted that sub-dicing is a very expensive process. The kerf must by necessity be small but is compromised by blade limits (typically a minimum is a factor of 1/20 dicing depth). In our experiments we use a sub-dice factor of 4 in both x and y, this involves about an order of magnitude increase in dicing/infiltration cost with susceptibility to failure (pillar toppling) an important factor.

Fig. 7 shows the response for the same 855 material prepared by the aqueous slip casting process, which in this instance achieved only a low pore density of ~50 pores/mm^2. Even at this low pore density there is a resemblance to the response obtained with sub-dicing, albeit that the performance is not adequate with such a low pore count.

Fig. 8 shows the pulse response obtained with a sample wafer with a pore density of ~1000 mm^{-2}. The improvement in performance is remarkable. It does not show the low frequency mode exhibited by the manufacturer supplied discs. Also, of note in all of these tests the pulser and oscilloscope settings are unchanged. That the amplitude of the material with 1000 mm^{-2} pore density is greater than those with low or no oriented pores is a somewhat unexpected result, and commensurate with that for sub-dicing.

V. Conclusions

A piezomaterial with a microstructure with uniaxially oriented pores should be particularly advantageous for 2D transducer array fabrication. The oriented pore microstructure corresponds to a 3-1 composite, the complement to the 1-3 composite connectivity of a dice and fill pillar array. Pillar array 1-3 connectivity in principle confines vibrational modes to the piezoelectric component, with each pillar surrounded by polymer. In the case of a 3-1 composite the piezoelectric component has connectivity extending in all 3 dimensions, but the vibrational modes nevertheless are effectively confined to the axis parallel to the pores because transverse modes are scattered by the array of voids.

We have developed a slip cast process for manufacturing PZT material with a high density of pores with diameter ~5 μm oriented normal to the face of the wafer. Pore densities up to 2000 mm^{-2} have been achieved.

Ultrasonic characterisation using pulse-response measurements provides a direct demonstration of unwanted mode suppression in the oriented pore transducer material compared to isotropic material. The results show that the oriented pore PZT wafers we produce can be used to fabricate 2D arrays in which undesirable vibrational modes are suppressed. This can normally only be achieved using conventional materials after very expensive sub-dicing processes.

Acknowledgment

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