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Experimental and simulated performance of lithium niobate 1–3 piezocomposites for 2 MHz non-destructive testing applications

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ABSTRACT

Lithium niobate piezocomposites have been investigated as the active element in high temperature resistant ultrasonic transducers for non-destructive testing applications up to 400 °C. Compared to a single piece of lithium niobate crystal they demonstrate shorter pulse length by 3×, elimination of lateral modes, and resistance to cracking. In a 1–3 connectivity piezocomposite for high temperature use (200–400 °C), lithium niobate pillars are embedded in a matrix of flexible high temperature sealant or high temperature cement.

In order to better understand the design principles and constraints for use of lithium niobate in piezocomposites experiments and modelling have been carried out. For this work the lithium niobate piezocomposites were investigated at room temperature so epoxy filler was used. 1–3 connectivity piezocomposite samples were prepared with z-cut lithium niobate, pillar width 0.3–0.6 mm, sample thickness 1–4 mm, pillar aspect ratio (pillar height/width) 3–6, volume fraction 30 and 45%. Operating frequency was 1–2 MHz.

Experimental measurements of impedance magnitude and resonance frequency were compared with 3-D finite element modelling using PZFlex. Resonance frequencies were predicted within 0.05 MHz and impedance magnitude within 2–5% for samples with pillar aspect ratio $\geqslant$3 for 45% volume fraction and pillar aspect ratio $\geqslant$6 for 30% volume fraction. Laser vibrometry of pulse excitation of piezocomposite samples in air showed that the lithium niobate pillars and the epoxy filler moved in phase. Experiment and simulation showed that the thickness mode coupling coefficient k_t of the piezocomposite was maintained at the lithium niobate bulk value of approximately 0.2 down to a volume fraction of 30%, consistent with calculations using the (Smith and Auld, 1991 [1]) model for piezocomposites.

1. Introduction

The purpose of this work was to investigate design principles for lithium niobate piezocomposites intended for use in ultrasonic transducers for non-destructive testing at high temperatures. Lithium niobate is a piezoelectric material with a high Curie temperature of 1210 °C, among the highest of the Pb-free ferroelectric materials [2]. In the form of a single crystal plate, lithium niobate has been used to make ultrasonic transducers capable of high temperature use for non-destructive testing and inspection of industrial plant during operation, both as single element transducers for scanning or continuous monitoring (at 400 °C [3–5] and most recently up to 1000 °C [6]) and as phased arrays [7] (including beam steering using an array controller and image reconstruction using data collected from individual elements at 400 °C). Development of lithium niobate 1–3 connectivity piezocomposites is a novel extension to this work and high temperature ultrasonic measurements have been demonstrated up to 400 °C, including initial verification of ultrasonic operation at high temperature on steel test objects [8], phased array operation at 2 MHz of eight adjacent $1 \times 10\,\mathrm{mm}^2$ array elements, centre to centre spacing 1.5 mm, with a commercial array controller [9,10], excitation of individual $10 \times 0.25\,\mathrm{mm}^2$ electrodes of a 36 element flexible array [11], and acoustic emission testing using $12 \times 12\,\mathrm{mm}^2$ piezocomposite samples [12]. This work was carried out using lithium niobate piezocomposites of 1–4 mm thickness, giving an operating frequency of 1–2 MHz. Acoustic emission signals were received mainly in the frequency range 0.1–1 MHz.

Use of piezocomposite materials is based on the principle that many piezoceramic materials such as lead zirconate titanate (PZT) show enhanced electromechanical properties when diced into pillars. The material can vibrate in the more efficient "bar mode" (coupling coefficient k_{33}) rather than the thickness mode which applies for a continuous plate (coupling coefficient k_t), thus

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giving an enhancement of transducer efficiency of 20–40% according to the Smith and Auld model [1]. In addition piezocomposites exhibit lower acoustic impedance (can be chosen in range 20–80% of the piezoelectric material, depending linearly on volume fraction of piezoelectric material) and broader bandwidth (improvement of 20–30% is possible) than solid plates, and flexibility in design is enabled because pillar dimensions and geometry and the properties of the passive material can all be changed. These advantages must be set against the extra manufacturing steps required. For lithium niobate k_{33} is not larger than k_t so an enhancement of transducer efficiency will not be expected. However we found that lithium niobate piezocomposites provided broad bandwidth (40–60%), with reduction of mechanical quality factor Q_m from 500 for z-cut material (found by fitting to the bulk material impedance response to the finite element model [13]) to 10 for piezocomposite, elimination of plate modes, and better resistance to cracking.

In this work we revert to epoxy filler, rather than high temperature filler, in order to examine more closely at room temperature the design principles and constraints of 1–3 piezocomposite lithium niobate transducer structures operating in the 1–2 MHz frequency range. z-cut lithium niobate was used, k_t = 0.17–0.23, in preference to y/36°, k_t = 0.49, because the properties of z-cut lithium niobate are more symmetrical around the *z*-axis, leading to a more symmetrical beam profile than Y/36° material. This has been observed through comparison of the beam profile produced by individual array elements fabricated on lithium niobate single crystal [14], in steered-beam directivity plots [7] (where single crystal z-cut lithium niobate was steered to 70° whilst retaining 50% amplitude, whilst Y/36° dropped to 10% amplitude by a steering angle of 50°) and for lithium niobate piezocomposite arrays in sector scan mode [9] where the strongest backwall reflection was offset by 5–10° from the axis for the Y/36° sample.

In this paper we look in more detail at the properties of lithium niobate piezocomposites in order to determine the design principles which will guide their use in high temperature transducers and arrays. We describe fabrication of piezocomposite samples with epoxy filler, analytical and finite element simulation, and experimental measurements of impedance magnitude vs frequency from which the piezocomposite coupling coefficient k_t was obtained. Results k_t were compared with values predicted using the calculations of Smith and Auld [1]. Laser vibrometry was used to confirm the mechanical mode of operation of the piezocomposite under pulse excitation in air.

2. Experimental and simulation procedures

2.1. Fabrication

The piezocomposites were fabricated by the 'dice and fill' method [15]. A single crystal plate of z-cut lithium niobate, dimensions $10 \times 10\ mm^2$, thickness 4 mm, was bonded using removable wax to a backing plate and two sets of parallel cuts were made at 90° to each other using a dicing saw to produce pillars with a square cross-section. The pillars were backfilled with Araldite CY1301/HY1300, a hard-setting epoxy commonly used in piezocomposites. Excess filler and undiced material were removed from the piezocomposite by precision lapping.

We fabricated a series of piezocomposite samples from an initial 4 mm thick lithium niobate plate, with lithium niobate pillar width 0.3–0.6 mm, pillar aspect ratio (PAR, pillar height/pillar width) of 3–6, and volume fraction (v.f.) of lithium niobate 30 and 45%. Nominal pillar width was 0.35 mm for 30% v.f. and 0.59 mm for 45% v.f. The same nominal kerf (saw cut width) of 0.29 mm was used throughout. Devices of the same volume fraction had the same pillar width. Pillar aspect ratio was altered by reducing the thickness of the piezocomposite by lapping, thus reducing the pillar height. Note that all piezocomposites of the same volume fraction originated from the same starting material. The piezocomposites were carefully inspected and found to be undamaged by the dicing and lapping process.

The frequency of operation was 1–2 MHz which falls in a suitable range for non-destructive testing applications. The two chosen volume fractions were considered sufficient to access the most appropriate pillar aspect ratios in our operating frequency range (i.e. just above the lower acceptable limit for 1–3 piezocomposites given by Smith and Auld [1]). In fact for a given sample thickness and a targeted range of pillar aspect ratio, a limited range of volume fraction is available for geometric reasons given the geometrical requirement for tall pillars and the practical requirements of manufacture. The width of the saw cut was maximised to allow the high temperature cement filler to be poured in between the pillars. The constraints which follow from this are: (1) for low volume fraction the pillars would have to be very narrow and could be too fragile; and (2) for high volume fraction, for the kerf to be narrow compared to the pillar size, the pillar aspect ratio would be too low. Impedance matching was not a significant factor in choosing the volume fraction since single crystal lithium niobate (Z = 36 MRayl) is already quite well matched to steel (Z = 45 MRayl) and for high temperature use there would be no requirement to lower the impedance to match to an aqueous couplant (Z = 1.5 MRayl).

Actual pillar size and kerf dimensions for use in the simulations were measured using a Veeco Dektak 3ST surface profiler on the top and bottom faces of the completed piezocomposite. Results are given in Table 1. Measurement uncertainty is 30 μm on pillar width and 10 μm on kerf. From these measurements it was deduced that the dicing process created square cross-section pillars with a slightly tapered shape, i.e. truncated square pyramids with the top approximately 10% narrower than the bottom. Overall, the pillars tended to be wider than the nominal dimensions, with narrower kerf. Samples were electroded by coating the entire top and bottom surfaces of the piezocomposite with silver paint (PELCO High Performance Silver Paste, Ted Pella, Inc.).

2.2. Simulation of piezocomposite performance

Lithium niobate belongs to the trigonal 3*m* class of crystalline piezoelectric materials (see e.g. Berlincourt et al. [16]). Therefore for a full three-dimensional model of the material's electromechanical behaviour, it must be fully characterised by six stiffness parameters at constant electric field c^E_{ij}, four piezoelectric stress coefficients e_{ij}, and two dielectric parameters at constant strain

Table 1
Average pillar and kerf width at top and bottom of pillars for lithium niobate/epoxy piezocomposites fabricated by dice and fill technique. Pillar aspect ratio (PAR) is pillar height/pillar width. Nominal pillar/kerf width was 0.35/0.29 mm for 30% v.f. and 0.59/0.29 mm for 45% v.f. Measurement uncertainty is 30 μm on pillar width and 10 μm on kerf.

	Pillar aspect ratio 6		Pillar aspect ratio 4		Pillar aspect ratio 3	
	Thickness (mm)	Pillar/kerf width (μm)	Thickness (mm)	Pillar/kerf width (μm)	Thickness (mm)	Pillar/kerf width (μm)
v.f. = 45%	3.54	629/271 725/172	2.36	680/207 714/179	1.77	653/228 710/176
v.f. = 30%	2.10	442/198 467/180	1.40	374/253 458/173	1.05	359/274 441/200