High temperature ultrasonic transducers using lithium niobate piezocomposite

Katherine J Kirk, Ruozhou Hou, Naga Mahesh Pragada, Louis Torbay, Nicole Schmarje and David Hutson
Microscale Sensors Group, School of Engineering, University of the West of Scotland, Paisley Campus, Paisley, Renfrewshire, PA1 2BE, UK
Telephone: +44 141 8483409
Fax: +44 141 8483663
E-mail: katherine.kirk@uws.ac.uk

Abstract

Lithium niobate piezocomposite transducers have been made for ultrasonic measurements up to 500°C. The piezocomposites were made by dicing y/36°-cut lithium niobate single crystal using an automatic dicing saw and filling with zirconia cement. Piezocomposite thickness was 1 mm, pillar width 0.4-0.8 mm, kerf width 0.5 mm, volume fraction of lithium niobate 30-40%. A transducer with an operating frequency 2.6-3.3 MHz was designed and fabricated with a steel case. This was used to demonstrate detection of artificial defects in a steel block, using high temperature couplant, up to 400°C. Stability of the high temperature properties and structure of lithium niobate was verified by impedance measurements and x-ray diffraction for lithium niobate single crystal and piezocomposite heated in air up to 500°C.

1. Introduction

Ultrasonic transducers for NDT typically use PZT piezoelectric material, and incorporate epoxy for bondlines, backing layers and as a passive material in piezocomposites. The transducers can typically tolerate 50-90°C but will not be able to withstand high temperatures unless actively cooled. Lithium niobate piezoelectric material has been proposed and used for high temperature transducers for ultrasonic testing at 400°C since 1989 (1). Lithium niobate piezocomposites were subsequently devised (2) in order to obtain improved mechanical and ultrasonic properties compared to an undiced plate of material, for example, broader bandwidth, shorter pulse length, lower $Q_m$, elimination of plate modes, and better resistance to cracking. The Curie temperature $T_c$ of lithium niobate is 1210°C, and the material is generally regarded as usable up to half $T_c$, i.e. 600°C, although very high temperature use of lithium niobate in ultrasonic testing up to 1000°C has recently been reported (3).

We previously demonstrated phased array operation of lithium niobate piezocomposite at 2 MHz using a commercial array controller (4,5), excitation of individual elements of a lithium niobate piezocomposite flexible array up to 250°C (6), and detection at high temperatures of acoustic emission test signals in the frequency range 0.1-1 MHz (7). Here we report our investigations on high temperature ultrasonic transducers made from lithium niobate 1-3 piezocomposites (i.e. pillars of piezoelectric material embedded in a passive matrix). We made stand-alone encased transducers which are fluid coupled to the test block using high temperature couplant and did not need to be clamped down. Unlike
our previous work the piezocomposite pillar arrays in the new transducers were made on an automatic programmeable dicing saw. Also in this work we focus on the properties of piezocomposite made from y/36°-cut lithium niobate material rather than z-cut material.

2. Modelling and analysis of y/36° lithium niobate piezocomposite transducers

Design of lithium niobate piezocomposites using y/36°-cut material was investigated at room temperature using epoxy filler for convenience. y/36° oriented lithium niobate plates, when electroded on the top and bottom faces, give the highest coupling coefficient available from lithium niobate for extensional mode waves although at the cost of symmetry in the beam profile. See references for comparison of beam profile of individual array elements on lithium niobate single crystal, steered-beam directivity plots and lithium niobate piezocomposite arrays in sector scan mode. We have shown how, for z-cut lithium niobate material, the Smith and Auld model does not predict the thickness mode coupling coefficient for a piezocomposite to rise above for a single crystal plate, unlike for PZT piezoceramic where transducer designers can obtain a 20-40% improvement. The Smith and Auld calculation for for y/36°-cut material would involve rotated equivalent materials parameters which were not available.

10 x 10 mm² piezocomposite samples were fabricated from an initial 4 mm thick lithium niobate plate by the ‘dice and fill’ method using a dicing saw (Model 15, diamond & wire disc saw, Logitech Ltd). The pillars were backfilled with Araldite CY1301/HY1300 hard-setting epoxy. The piezocomposites had 30% volume fraction (v.f.), pillar width 0.35 mm, kerf width 0.29 mm, and pillar aspect ratio (PAR, pillar height / pillar width) 3, 4 and 6. Note that samples of different PAR originated from the same starting material and pillar height was reduced by lapping. Nominal dimensions and expected resonance frequencies are shown in Table 1.

<table>
<thead>
<tr>
<th>PAR 3</th>
<th>PAR 4</th>
<th>PAR 6</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thickness (mm)</td>
<td>Expected resonance (MHz)</td>
<td>Thickness (mm)</td>
</tr>
<tr>
<td>1.05</td>
<td>3.1</td>
<td>1.40</td>
</tr>
</tbody>
</table>

Table 1: Nominal dimensions and resonance frequency for lithium niobate/epoxy piezocomposite with 30% v.f., pillar width 0.35 mm, kerf width 0.29 mm and pillar aspect ratio (PAR) 3, 4, 5.

Experimental impedance magnitude and phase results were compared with simulation by finite element modelling using PZFlex (Weidlinger Associates Inc.), Figure 1. We simulated a full pillar unit cell of average measured dimensions from Table 2 with translational boundary conditions. The full three-dimensional set of elastic, electrical, and electromechanical parameters for lithium niobate were used. The PAR 6 and PAR 4 samples show a well behaved response with a single clear thickness mode, although the PAR 4 experimental and simulated resonance frequencies do not match well. Resonance in the PAR 3 piezocomposite is substantially less pronounced than the simulated results.
Figure 1: Simulated and experimental electrical impedance magnitude and phase for y/36°-cut lithium niobate piezocomposites with PAR (a) 3, (b) 4, (c) 6.

The difference between experimental and simulated values cannot be explained by the experimental error in measurement, but could be due to problems of chipping during dicing the y/36°-cut material as the Logitech Model 15 saw was not ideal for the purpose. The PAR 3 piezocomposite may be more easily affected by inaccuracies in pillar width arising in manufacture. A similar discrepancy at low PAR and low volume fraction was
seen for z-cut lithium niobate piezocomposite (10) although this case was more difficult to explain as the piezocomposite pillars appeared intact.

<table>
<thead>
<tr>
<th>Measured dimensions (µm)</th>
<th>PAR 3</th>
<th>PAR 4</th>
<th>PAR 6</th>
</tr>
</thead>
<tbody>
<tr>
<td>top</td>
<td>pillar width</td>
<td>kerf width</td>
<td>pillar width</td>
</tr>
<tr>
<td>top</td>
<td>440</td>
<td>216</td>
<td>438</td>
</tr>
<tr>
<td>bottom</td>
<td>462</td>
<td>178</td>
<td>471</td>
</tr>
<tr>
<td>σ</td>
<td>31</td>
<td>11</td>
<td>34</td>
</tr>
</tbody>
</table>

Table 2: Average measured dimensions (µm) of y/36°-cut, 30% v.f. piezocomposites (standard deviation σ) showing the samples have square tapered pillars and quite a large variation in pillar size.

The key parameter of interest is the coupling coefficient of the piezocomposite $k_t$ which is obtained from the impedance magnitude curves using the series and parallel resonance frequencies $f_s$ and $f_p$ (Equation 1).

$$k_t^2 = \frac{\pi}{2} \frac{f_s}{f_p} \tan \left( \frac{\pi}{2} \frac{f_s}{f_p} \right).$$

Experimental and simulated $k_t$ values are compared in Figure 2. The form of the experimental impedance curve for PAR 3 does not allow calculation of $k_t$. The standard value for bulk lithium niobate of y/36°-cut orientation is 0.49 (15,16).

![Figure 2: Electromechanical coupling coefficient $k_t$ vs PAR from experimental and simulated impedance results for y/36°-cut lithium niobate piezocomposite, 30% v.f.](image)

Bandwidth results come from transmit/receive tests conducted using a DPR300 ultrasonic pulser–receiver (JSR Ultrasound, now Imaginant Inc.) with the piezocomposites clamped onto a 37 mm thick mild steel block. The return signal was low pass filtered with a cut off frequency of 10 MHz and the first back wall echo was fast fourier transformed. Figure 3 shows (a) the 6 dB bandwidth and (b) bandwidth normalised to the centre frequency (fractional bandwidth), for y/36°-cut piezocomposite compared to z-cut piezocomposite.
of two different volume fractions, 30 and 45%. Bandwidth obtained from the y/36°-cut samples corresponds qualitatively with the electrical impedance results, i.e. the y/36°-cut PAR 3 sample showed a flat but very wide peak in impedance, and as the pillar aspect ratio increased the peak narrowed. Overall, y/36°-cut lithium niobate piezocomposites were expected to have a broader bandwidth than z-cut piezocomposites due to a higher $k_t$, however this was not found at higher PAR.

![Figure 3: Bandwidth vs function of PAR for y/36°-cut (v.f. 30%) and z-cut (v.f. 30, 45%) lithium niobate piezocomposite. (a) bandwidth in MHz (b) normalised to centre frequency. Note that the centre frequency decreases with PAR.](image)

3. Manufacture of high temperature lithium niobate /cement piezocomposite samples

**Dicing:** A y/36° oriented lithium niobate crystal, starting dimensions 13 x 13 x 1.5 mm$^3$, was diced using an automatic dicing saw (DAD 3230, Disco Corp.) specified with a depth accuracy of ±1µm and positioning accuracy of ±3µm. Saw cut depth was 1 mm. Saw blade thickness 0.5 mm was used, with the gap between the pillars (kerf) expected to be slightly larger. Maximum available kerf width was used to facilitate adding the passive material in between the pillars. These dimensions give a volume fraction of lithium niobate 30-40% and PAR 1-1.3. Diced lithium niobate crystal is shown in Figure 4(a).

![Figure 4: (a) Diced lithium niobate samples, 13 x 13 mm$^2$ (b) lithium niobate piezocomposite with alumina cement, (c) piezocomposite with zirconia cement.](image)

**Filling:** Two high temperature ceramic adhesives / potting compounds were tried for the piezocomposite filler material. Both are specified to more than 1000°C and comprised a powder and a liquid binder which are mixed to a paste. Initially, alumina cement
Durapot 801, Cotronics Corp.) was tried as a passive material but unfortunately it degraded in water and tended to disintegrate during lapping, Figure 4(b). Zirconia cement (Resbond 940, Cotronics Corp.) proved to be a much better passive material, Figure 4(c). The diced sample with lithium niobate pillars was filled from the front face with zirconia based cement then placed in a vacuum oven. A vacuum created in the oven and then released helps push the cement into the kerfs to fill the sample properly. The oven temperature was raised to 50°C and the sample left overnight to cure.

**Lapping:** Excess lithium niobate and filler material on the top and bottom surfaces was removed by lapping to the desired final sample thickness using an aqueous suspension of 9 µm aluminium oxide powder as the grinding medium. The sample was mounted for lapping onto a glass sample holder with removable wax (melting temperature about 60°C).

### 4. High temperature tests on lithium niobate single crystal and piezocomposite

#### 4.1 X-ray diffraction

Lithium niobate single crystal and a piezocomposite sample were heated from room temperature to 500°C in 100°C steps. After each annealing step the sample was cooled and the x-ray diffraction pattern was captured. In Figure 5(a) two peaks were observed at 23.8° and 48.5°. These results show that for temperatures from room temperature until 500°C, the structure of the y/36° cut lithium niobate remained stable. The x-ray diffraction pattern for the lithium niobate piezocomposite is shown in Figure 5(b), focusing on the peak at 48°, where there was a small change on heating from room temperature to 100-200°C, but otherwise no significant change.

![X-ray diffraction pattern](image)

**Figure 5:** (a) X-ray diffraction pattern for lithium niobate single crystal from room temperature to 500°C. Measurements taken after cooling down from each temperature. (b) Detail of small peak at 48° for piezocomposite.

#### 4.2 Electrical impedance

Lithium niobate single crystal and lithium niobate piezocomposite samples were heated on a hot plate to 500°C in steps of 100°C. Samples were electroded on front and back
faces with silver paint (PELCO High Performance Silver Paste, Ted Pella, Inc) which was rated up to 900°C and found to perform well in our application up to 500°C. At room temperature the resonance frequency and the anti-resonance frequency for the lithium niobate single crystal were 3.2 and 3.6 MHz respectively (Figure 6). Impedance magnitude measurements for (a) lithium niobate single crystal and (b) piezocomposite, measured at high temperature, are shown in Figure 7. It can be seen that the piezocomposite exhibits fewer lateral modes at low frequency than the single crystal sample. On heating up to 500°C the resonance frequency for lithium niobate single crystal remained the same within 0.1 MHz, and within 0.2 MHz for lithium niobate piezocomposite up to 400°C.

![Impedance magnitude and phase](image)

**Figure 6: Lithium niobate single crystal at room temperature (a) impedance magnitude, (b) impedance phase.**

The peak values of impedance magnitude for single crystal and piezocomposite samples at different elevated temperatures up to 400-500°C, extracted from the graphs of Figure 7, are shown in Figure 8. It is interesting to note that the peak impedance of lithium niobate piezocomposite shows the opposite trend with temperature to single crystal material. In both cases the electromechanical coupling coefficient $k_t$ remained constant within 3% during heating.

In another experiment electromechanical coupling coefficient $k_t$ of two piezocomposite samples was measured at room temperature after heating on the hot plate at 100°C intervals up to 500°C. Both samples were 12 x 12 x 1 mm$^3$, pillar width 0.8 mm, pitch 1.3 mm, volume fraction 38%. Results are shown in Figure 9. $k_t$ of Sample 1 remained constant within 6% but Sample 2 cracked on cooling from 300°C causing a large reduction in $k_t$. The cause of the cracking is believed to be build up of stress on cooling due to the pyroelectric effect which is present in lithium niobate. (Note that the thermal expansion coefficient of lithium niobate $15.4 \times 10^6$ °C$^{-1}$ parallel to z-axis in a temperature range from 0°C to 110°C is very similar to that of steel.)
Figure 7: Impedance magnitude vs frequency measured at temperatures up to 500°C (a) single crystal lithium niobate (b) lithium niobate piezocomposite.

Figure 8: Peak impedance magnitude vs temperature for lithium niobate single crystal and lithium niobate piezocomposite.
Figure 9: Coupling coefficient vs annealing temperature for two lithium niobate piezocomposite samples, heated on a hot plate and measured after cooling from each temperature. Sample 2 cracked on cool down from 300°C.

5. High temperature transducer

5.1 Manufacture and testing

The lithium niobate piezocomposite transducer for high temperature ultrasonic testing was designed with an operating frequency range of 2.6-3.3 MHz. A lithium niobate single crystal plate was diced into pillars and filled with zirconia cement. The acoustic impedance of lithium niobate and steel are closely matched at 35 and 45 MRayl respectively, therefore there is no need for a matching layer. After curing and electroding the piezocomposite, the device was fabricated with a steel case and the same zirconia cement used as a backing layer. Piezocomposite characteristic dimensions were 33% volume fraction, 1 mm pillar height, 0.8 mm pillar width, giving PAR 1.3. Overall dimensions of the piezocomposite used to fabricate the transducer were 13 x 13 x 1 mm³. Figure 10 shows the transducer design, and Figure 11(a) shows the finished transducer.

Figure 10: Design of transducer for high temperature ultrasonic testing.

From the impedance magnitude characteristic in Figure 11(b) we can obtain the series and parallel resonance frequencies 2.67 and 3.14 MHz respectively (compared to the expected value of 3.69 MHz). The electromechanical coupling factor $k_t$ of the transducer
was calculated from Equation 1 to be 0.56. The operating frequency of the transducer calculated from two successive peaks of the backwall echo was 3.2 MHz.

![Graph](image1)

**Figure 11:** (a) photograph of finished transducer, (b) impedance magnitude.

### 5.2 Pulse-echo testing

The transducer was tested on a 30 mm thick steel block, longitudinal sound velocity 5980 ms⁻¹, with two side drilled holes at 10.7 and 16.8 mm depth below the top surface, Figure 12(a, b). Pulse-echo results at room temperature using Soundsafe couplant (Diagnostic Sonar Ltd) show detection of the 16.8 mm depth hole at 7 and 17 µs, Figure 12(c,d). The first 3 µs of the main bang is not shown.

![Graph](image2)

**Figure 12:** (a,b) Pulse-echo testing setup on hotplate for 30 mm steel block with side drilled holes (a) backwall only, (b) detection of hole at 10.7 mm depth, (c,d) room temperature pulse-echo response (c) backwall, (d) hole at 16.8 mm depth detected at 7 and 17 µs. First 3 µs of main bang not shown.
Three couplants were chosen for use at different temperatures. Soundsafe couplant can withstand 140°C and was used at 100°C. At higher temperatures an ‘anti-sieze’ copper-based grease was used. Although this is not an official high temperature couplant it worked in the range 200-300°C. For the target temperature of 400°C Sono 900 ultrasonic couplant (Sonotech Inc) was used. This is a thick gritty paste specified for 315-480°C. We found that this performed well and produced less fumes than Sono 1200 (specified for 370-600°C) which was also tested. For high temperature measurements the hot plate was heated to 400°C. Figure 13 shows detection of the hole at 10.7 mm depth, 3.6 µs after the first backwall echo. The result shows that the transducer was in good operational condition at high temperature.

Figure 13: Pulse-echo response of piezocomposite transducer on steel block at 400°C (a) no flaw, (b) detection of hole, 10.7 mm depth, 3.6 µs after first backwall echo.

6. Conclusions

Lithium niobate 1-3 piezocomposites with cement filler material were fabricated and tested at high temperatures by impedance measurements and x-ray diffraction. A high temperature transducer was constructed using a piezocomposite sample, and pulse-echo tests were carried out on a steel block heated on a hot plate. Using high temperature couplant it was possible to detect a side-drilled hole in the block at 400°C. A high piezocomposite thickness mode coupling coefficient $k_t$ of 0.50-0.55 was obtained from the lithium niobate / cement piezocomposites, similar to the standard bulk value of 0.49, for samples with volume fraction 30-40% and pillar aspect ratio (PAR) 1-1.3.

To explore the principles for transducer design with y/36°-cut lithium niobate, piezocomposites with PAR 3, 4 and 5 were made using epoxy filler. Experimental impedance measurements were found to match well with 3-D finite element modelling for PAR 4 and 6, with a single thickness mode resonance obtained. However a rather broad and flat resonance was observed experimentally for PAR 3 samples, not predicted in the model. The effects observed in the (small signal) impedance measurements were confirmed by bandwidth measurements from transmit-receive tests using an ultrasonic pulser-receiver.
We conclude that lithium niobate piezocomposite can be used to make a stand-alone high temperature transducer for thickness measurement and flaw detection. The piezocomposite material and the lithium niobate itself showed no deterioration at high temperatures. Although a higher PAR would generally be preferred for piezocomposites, the lithium niobate / cement samples showed a clear resonance mode at the expected frequency and satisfactory performance with PAR 1-1.3.

7. References

10. K.J. Kirk, N. Schmarje, ‘Experimental and simulated performance of lithium niobate 1–3 piezocomposites for 2 MHz non-destructive testing applications’, Ultrasonics, http://dx.doi.org/10.1016/j.ultras.2012.05.007