Metal-electroceramic bonding in PZT through the selective application of laser energy

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Ferrite powder has been directly bonded to the surface of poled lead zirconate titanate (PZT-4) using direct laser sintering. The resultant cohesion between the metal and ceramic is extremely good although care must be taken with the processing in order to limit the damage inflicted upon the PZT. Four point bending suggests that the strength of the processed samples has reduced to 60 MPa as compared to 76 MPa for the unprocessed ceramic. Electrical and piezoelectric measurements shows that the laser sintering had caused mechanical damage to a depth of 550 μm and thermal damage (depoling) to a depth of 800 μm.

1. Introduction

Since their invention, piezoelectric materials based on lead zirconate titanate (PZT) have become commonplace in a large array of applications such as spark igniters, vibration control, ultrasound and sonar [1–3].

In order to use a piezoelectric material as an actuator material, a means must be provided by which (a) an electric field may be applied across the ceramic and (b) the mechanical force and displacement generated by the ceramic can be transmitted usefully into another structure. A conventional means to provide an electrode may be the use of a fired on paint, such as a metal loaded glass which diffusion bonds to the ceramic with the application of heat. The electroded, poled, ceramic would then be bonded to some sort of structure, typically using a conventional adhesive such as an epoxy.

This adhesive layer can form a weak link between the piezoelectric and the structure to be actuated, due to significant signal absorption and loss of frequency response [4]. This is as a result of, among other things, the poor impedance match between the ceramic and a polymeric adhesive [5, 6]. A more efficient way to couple to the piezoelectric to a structure would be to embed the piezoelectric directly onto or into the structure to be actuated. One would expect this to lead to excellent sensitivity and response time. If allied to the introduction of a compressive stress in the ceramic, one would also expect to see significant improvements in reliability.

A number of methods of bonding metal to electroceramics have been previously reported on: active brazing [7], using pure metal pastes as the bonding medium [8], and low temperature sintering [9], although the active brazing approach suffers from unwanted reactions in the electroceramic, and the pure metal paste approach is expensive. All of these techniques are limited in practice to relatively simple geometries, and so do not lend themselves to the development of smart net shape products with embedded sensing and actuating potential.

This paper reports on research which has focused on examining an alternative approach. The overall aim would be to use selective laser sintering techniques to fabricate a component around an electroceramic, such that products of any geometry could be created with an embedded sensor/actuator. This approach would build upon laser bonding techniques [10] to encapsulate the actuator. The first stage in developing a product in this way is to examine whether a scanning laser can be used to generate a bond between metal powder and an electroceramic substrate,

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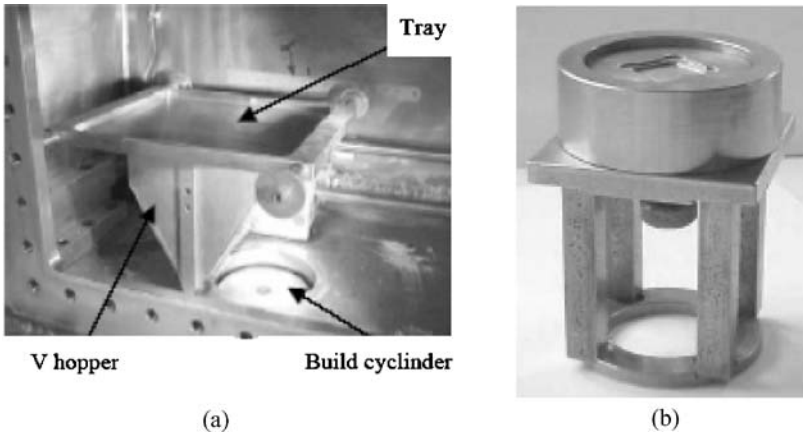

Figure 1 Schematic of sinterstation.

and that examination was the aim of the work presented here.

2. Materials and equipment

The electroceramic used in this study was PZT-4 (lead zirconate titanate), with samples typically 20–25 mm diameter or square and 2 mm thick. The metal powder used was ferrite, with an average particle diameter of 60 μm. Ferrite powder was employed as it is a simple non-alloyed "model material", in order to try and understand the processing; more complicated materials such as aluminium alloys are often employed using laser sintering but were not studied here. An experimental sinterstation, fabricated at the University of Leeds was used to carry out all laser scanning experiments. This has been described in full elsewhere [11], and so is only briefly described here. The machine is shown schematically in Fig. 1, and uses a 240 W CO_2 laser, with a 1.1 mm diameter spot size at the bed. For experiments using only metal powder the powder was contained in a tray, shown in Fig. 2a, where attempts were being made to bond the metal powder to a PZT substrate the PZT substrate was held in the the jig shown in Fig. 2b, which located in the build cylinder shown in Fig. 2a, and with a controlled depth of powder spread above the PZT. All of the experiments reported here were carried out in an argon atmosphere. The PZT substrate (typical diameter of 20 mm) was located in the square recess in Fig. 2b.

3. Experimental characterisation of powder/laser interaction

Initial experiments focused on developing an understanding of the laser scanning parameters which would cause the ferrite powder to melt/sinter. These experiments were carried out using single linear scans of the laser beam onto the powder using a variety of laser powers and scanning speeds. The results of a typical set of experiments can be seen in Fig. 3. Good sintering was defined as producing tracks of a high quality, repeatability and uniformity. Fig. 4 shows an overall process map which was built up for single scans of the material, and also indicates the range of laser powers and scan speeds which were used. Good sintering was seen at laser powers of 5–45 W, with scan speeds of 1–5 mm/s. Tracks produced at these conditions had a measured depth of 0.4–0.9 mm. Full details of the process mapping process can be found in [11].

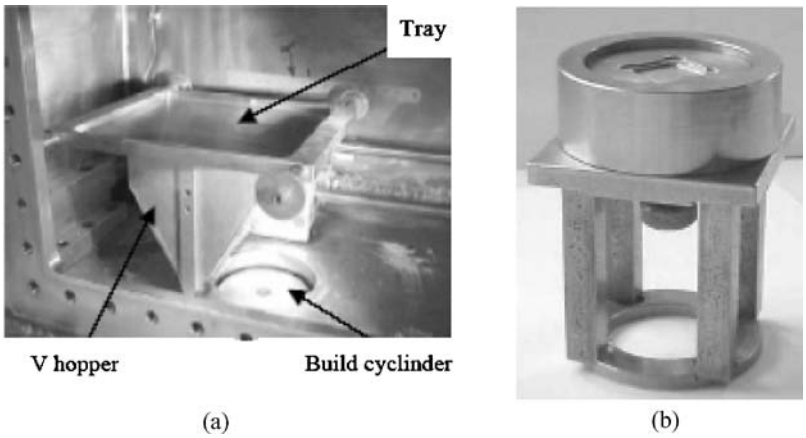

Figure 2 (a) Tray in build chamber, (b) Jig for bonding trials.

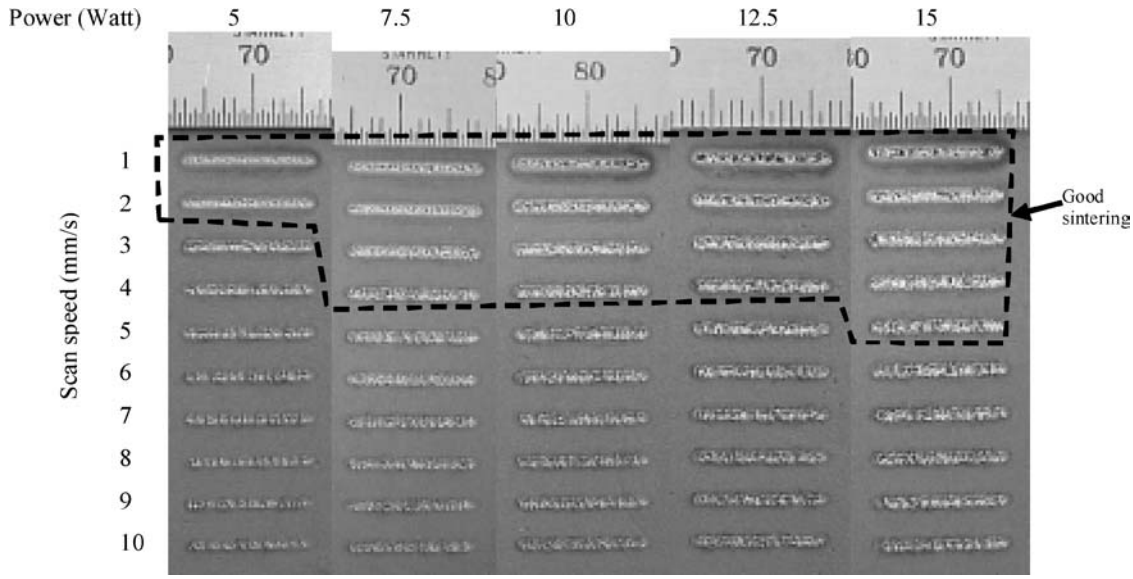

Figure 3 Tracks of ferrite powder. Single line scanned in an argon atmosphere.

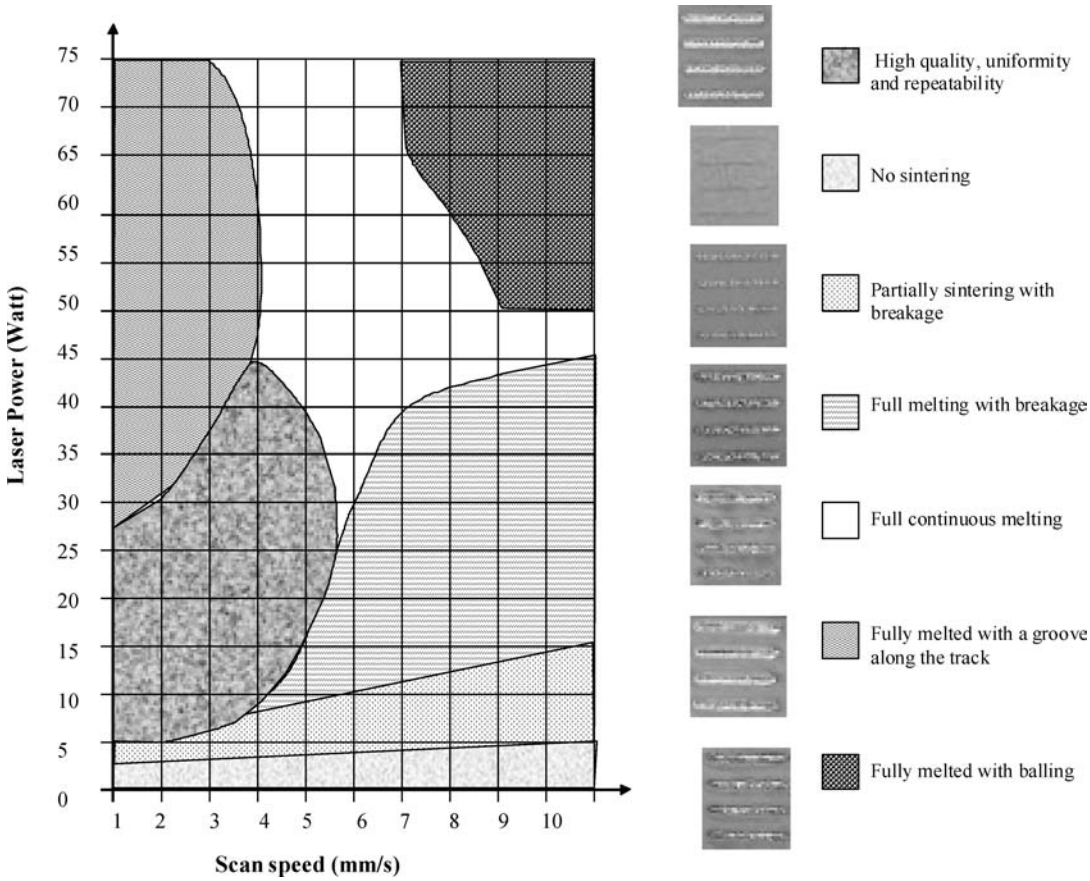

Figure 4 Process map for ferrite powder tracks. Single line scanned in an argon atmosphere.

4. Experimental bonding trials and assessment

4.1. Single line scans of ferrite powder with PZT substrate

In the initial trials which aimed at generating bonding between ferrite and PZT using a scanning laser beam the approach used was simply to use the jig shown in Fig. 2b, with a 0.4 mm depth of ferrite powder spread evenly above the substrate, and to use single line scanning to both melt the ferrite and bond it to the PZT substrate below. Fig. 5 shows a cross section of one experiment of this form, with SEM images also shown in Fig. 6. In both cases the unscanned powder has been removed. Both figures show that the electroceramic has cracked during the process, as a result of thermal shock as the laser energy has been

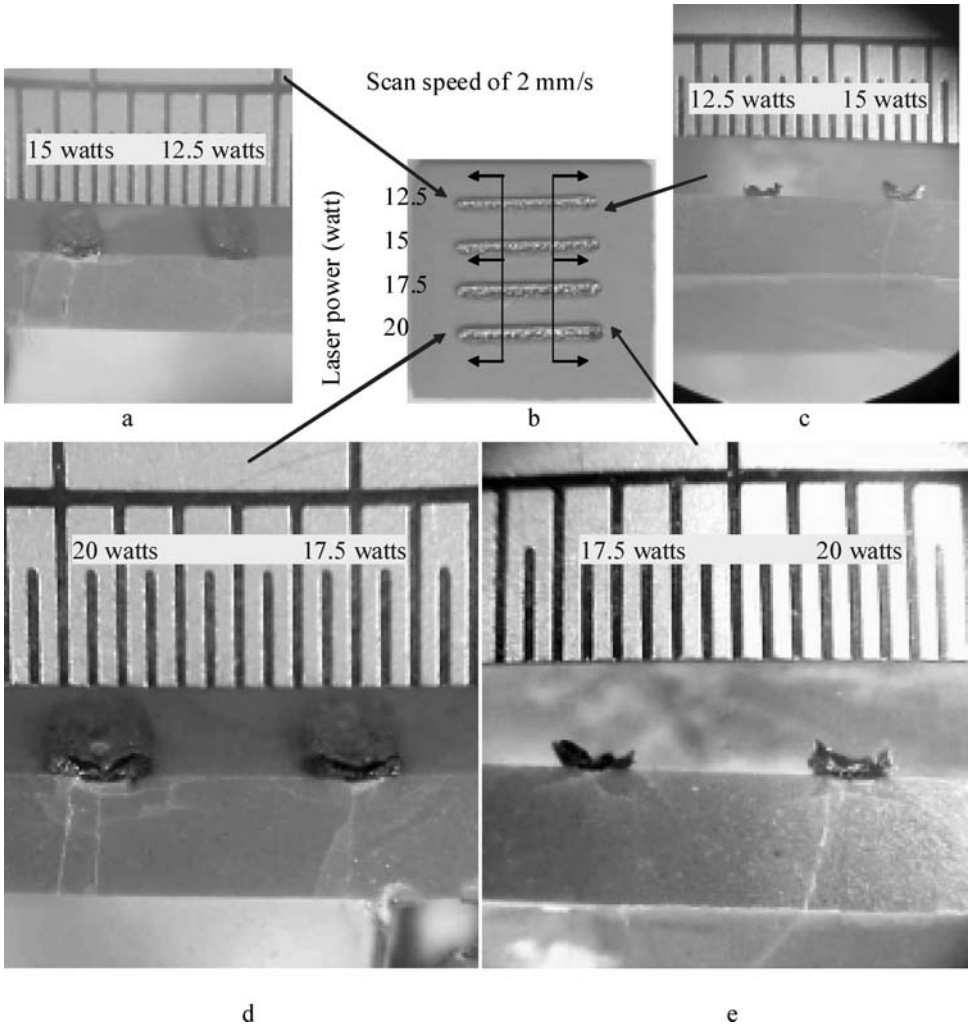

Figure 5 Single line scans of ferrite on PZT. Argon atmosphere.

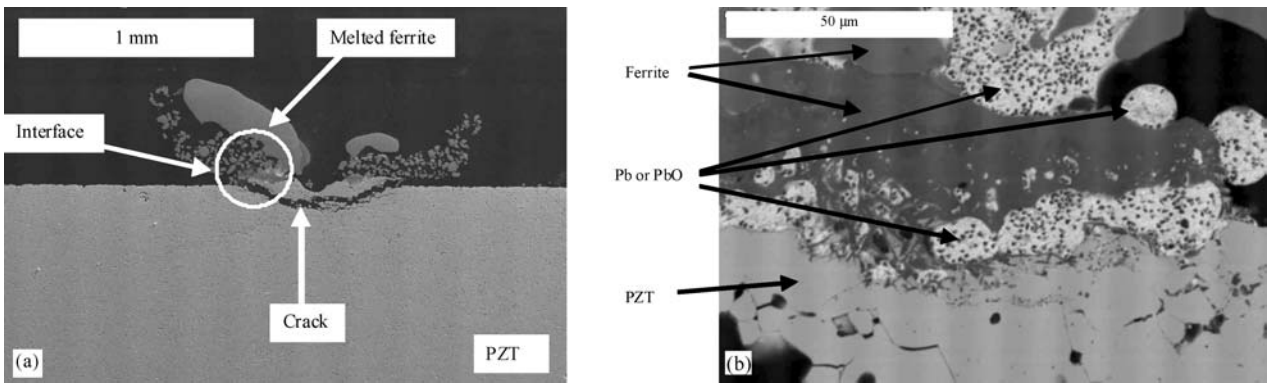

Figure 6 (a) SEM of left hand scan from Fig. 5e; (b) SEM backscattered image of area circled in Fig. 6a.

transferred to the ceramic. However, the images also show some positive outcomes from this series of tests: the ferrite powder has consolidated well, and there is evidence of reaction bonding at the interface, illustrated most clearly in Fig. 6b where the lead or lead oxide can be seen to have segregated from the PZT to form an interface between the PZT and the ferrite. There is no evidence of any wetting problems at the interface. Overall these results suggested that if the thermal shock on the PZT could be eliminated that a strong bond between the ferrite and the PZT would be established.

The obvious way to reduce the thermal shock on the PZT is to reduce the laser power, and it was experimentally determined that in order to avoid cracking in the PZT laser powers below 12 W were required at scan speeds between 2 and 5 mm/s. However, experiments at lower

laser powers, whilst not cracking the PZT, either did not sufficiently melt the ferrite for consolidation to occur, or did melt the ferrite but did not generate any bonding between the ferrite and the PZT. Consideration of Fig. 4 with this information indicates that additional energy is being required with the PZT substrate in place in order to drive the bonding reaction.

4.2. Repeated line scanning of ferrite powder

In order to reduce the thermal shock on the PZT a method a pre-heating the PZT was required. A number of alternative approaches were considered, however, within the confines of a feasibility study it was decided that the most effective way of evaluating the effect of preheating was to pre-heat through scanning the powder a number of times at a lower power rather than scanning once at the laser powers identified in Fig. 5. To investigate this approach a number of experiments were carried out where repeated line scans of the ferrite powder only were used to characterize the impact this would have on the scanning conditions. This series of experiments examined the influence of the laser power, scan speed, and number of repeat scans on the consolidation of the ferrite powder. In all cases the repetition happened in the same scanning direction, with the repeat scan following 0.22 s from the end of the previous scan; the total time between scanning at the same point varied from 0.81 s (scanning at 17 mm/s) to 5.22 s (scanning at 2 mm/s). Fig. 7 shows a typical set

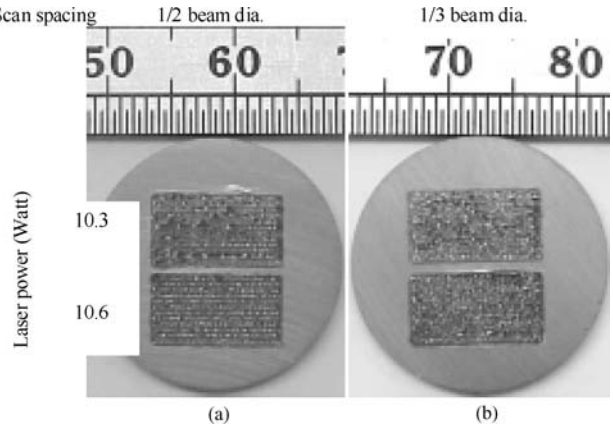

Figure 8 Line scans of ferrite on PZT. Each line scanned twice at a scan speed of 2 mm/s.

of results from this process, with the definition of good sintering the same as for Section 3.

Comparing Fig. 7 with Fig. 3 shows that repeat scanning does change the processing window, as, for a given power level, the range of scan speeds at which good sintering occurs has been moved upwards. Overall it was found that repeating the scanning up to four times had a significant impact on the extent to which a track would consolidate, but that beyond four repetitions the impact of the additional scans became marginal. In addition the thickness of the tracks reduced: those shown in Fig. 7 had a measured depth of 0.2–0.4 mm.

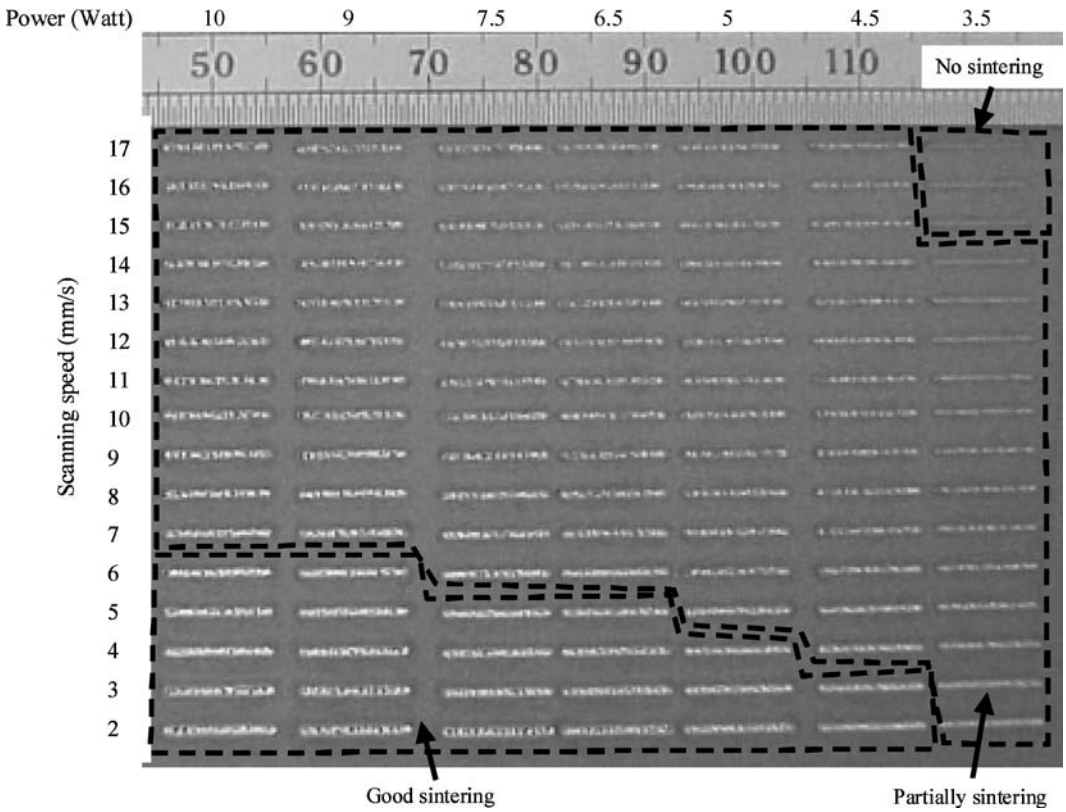

Figure 7 Tracks of ferrite powder. Each line scanned four times at the power/speed indicated. Argon atmosphere.

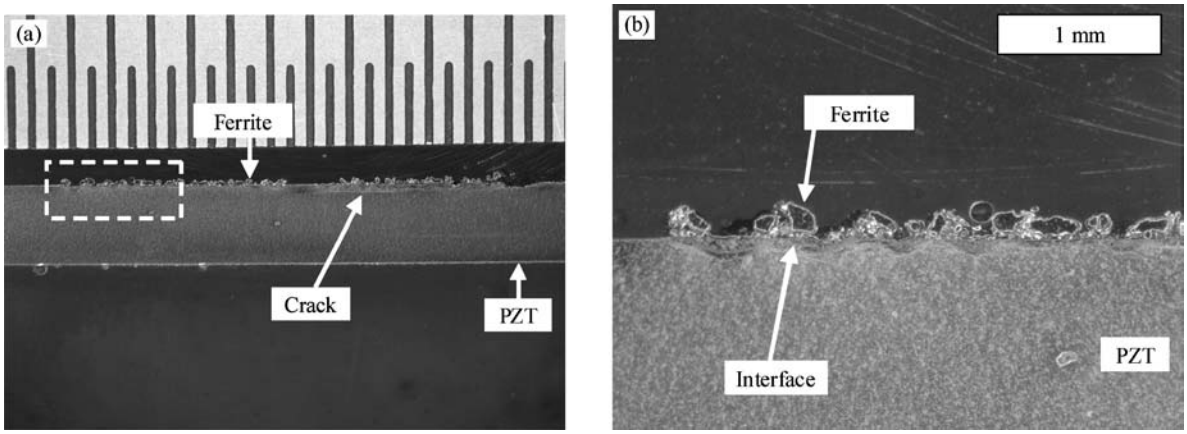

Figure 9 (a) Cross section of repeated single scans of ferrite on PZT, (b) Magnified view of boxed area in Fig. 9a. Processing conditions as for Fig. 8b.

4.3. Repeated line scans of ferrite powder with pzt substrate

Figs 8 and 9 illustrate the results of the repeated line scanning approach when applied with a PZT substrate. For all the experiments reported in this section the depth of powder spread above the substrate was 0.15 mm. As is shown in Fig. 8 the experiments proved successful enough for layers of material rather than just lines to be deposited. The scanning conditions for those samples shown in Fig. 8 were that each line was scanned twice, with the same start point and 0.22 s delay between the end of a scan and the start of its repetition. The next scan line was then offset by an amount known as the scan spacing, so in Fig. 8a the scan was offset by one half of the laser spot size, and in Fig. 8b the scan was offset by one third of the laser spot size. The relatively small offset means that either one half or two-thirds of the material in a track is scanned again when the adjacent track is scanned, effectively replicating the repeat line scan process for a significant amount of material in the original track.

Fig. 9a shows that the repeat scan process did not completely eradicate the cracking seen when single line scans were used. However, Fig. 9b shows a very well defined and crack free interface layer between the PZT and the ferrite.

SEM of these samples confirmed that the interface was as suggested by Fig. 6b, with an interfacial compound formed through mixing of the ferrite and the PZT, an interaction which causes the lead or lead oxide to segregate out. The interface depth was a fairly constant 0.1 mm. Energy dispersive x-ray analysis across the interface indicated that at points in the interface close to the PZT about 25% by weight of the material present was Fe, and that in the interface area away from the PZT about 70% by weight of the material present was Fe [11], indicating a gradual change across the 0.1 mm depth of interface from Fe to PZT.

4.4. Bending tests

In order to gain an initial mechanical assessment of the mechanical properties of the interface a series of bending tests were carried out on samples produced using the conditions which produced the sample shown in Fig. 8b. Four point bend testing was used, with the test set-up as shown in Fig. 10a and b. Dimension L was 13 mm, a was 2.75 mm, with h 2.2 $\pm$ 0.1 mm and b in the range 1.5–2 mm. The ferrite layer thickness was in the range 0.18–0.36 mm. Eight samples were tested in all, in the orientation shown, with the ferrite layer on the tension side (worst case for delamination), and all eight samples failed in the manner shown by Fig. 10c and d. In all cases the failure initiation point was between the two inner supports, making the test valid, and in all cases the failure initiation point was within the PZT, approximately 0.5 mm away from the interface.

Over the eight tests the average failure load was 30 N (range 23 N to 41 N), which roughly equates to an average maximum stress in the specimen of 60 MN/m^2. The quoted maximum tensile strength of PZT-4 is 11,000 PSI (76 MN/m^2) [12].

4.5. Electrical and piezoelectric measurements

Small rectangular samples were cut from laser processed samples using a diamond saw. The specimens were then systematically thinned from the treated electrode side to progressively remove material affected by the laser sintering, always leaving the back original electrode intact. The PZT materials used were poled prior to laser processing.

d_{33} and the relative permittivity was measured for the progressively thinned specimens in conjunction with unprocessed control materials. Values for d_{33} and the relative permittivity were collected using a piezo-meter (Piezotest). Figs 12–13 show the effect of thinning the materials on the d_{33} and relative permittivity respectively. For each data point, an average of five samples was used for both the processed and control materials.

After removing ca. 800 μm of material, d_{33} returns to the same value as for the control samples. Removing just ca. 550 μm of material shows a recovery of the relative permittivity. This may be explained by the type of damage

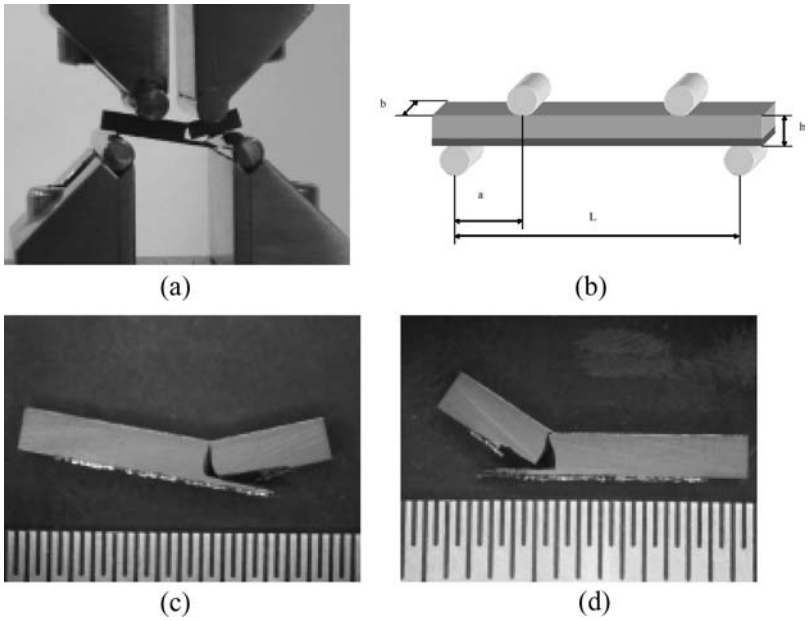

Figure 10 Bend test: (a) & (b) Arrangement, (c) & (d) Outcomes.

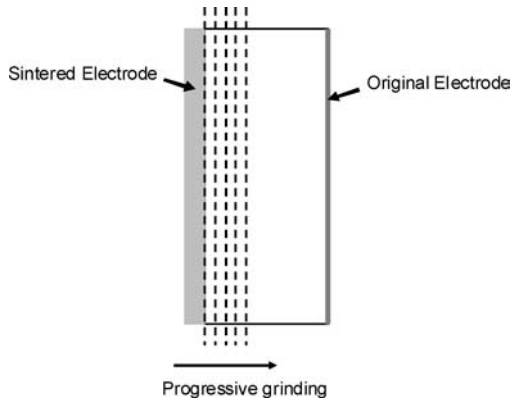

Figure 11 Representation of progressive thinning technique used for electrical and piezoelectric analysis.

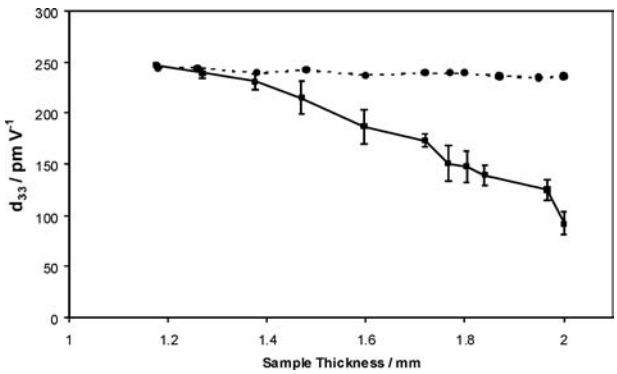

Figure 12 d_{33} as a function of thickness as poled laser sintered samples are progressively thinned. The dashed line shows data for the control samples.

inflicted during laser sintering. Thermal shock or heating to a temperature near the transition temperature (the temperature above which the piezoelectric effect no longer exists in a particular material, 320°C in the case of the

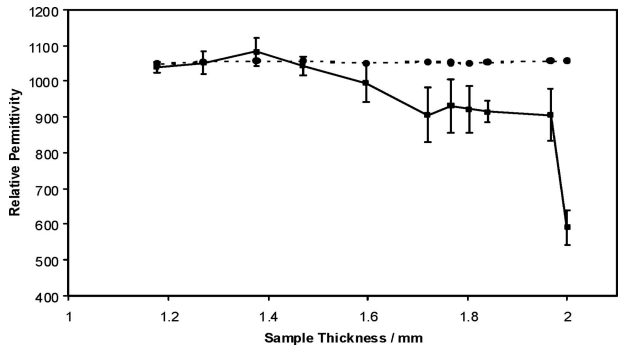

Figure 13 Relative permittivity as a function of thickness as poled laser sintered samples are progressively thinned. The dashed line shows data for the control samples.

piezoelectric 4D) will be sufficient to create drop in d_{33}. The relative permittivity will also be affected slightly by depoling in these materials, but to provide the large scale reduction that is observed, physical damage must be inflicted, be it chemical or physical. The chemical damage could be in the form of, for example, PbO loss leading to a change in composition. The physical damage would be from cracking, whereby air gaps are introduced into the structure, leading to an overall drop in bulk relative permittivity. Interestingly, after the first thinning process, which involved simply removing the laser sintered electrode plus a minute quantity of ceramic, the largest improvement in relative permittivity occurs.

5. Conclusions

1. The use of laser scanning of metal powders to generate metal-electroceramic bonding has been demonstrated to be feasible, but requires very careful process control to produce a good quality bond whilst retaining the integrity of the electroceramic.

2. Repeat scanning has been shown to offer a mechanism by which the processing window in the laser scanning of powder beds can be enhanced by reducing damage due to thermal shock.

3. The damage developed in the ceramic by the use of laser sintering has been characterised by the use of mechanical, electrical and piezoelectric analysis.

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