KES Transactions on Sustainable Design and Manufacturing I
Sustainable Design and Manufacturing 2014 : pp.950-960 : Paper sdm14-097

Laser transmission welding of polymers and ceramics demonstrated on PMMA and LTCC

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Abstract Joining technologies for incompatible materials are of high interest in many application areas as they allow the combination of the advantages of different materials. This study demonstrates a joining technique for transparent polymers and ceramics using commonly applied materials as examples, namely polymethylmethacrylate (PMMA) and low-temperature co-fired ceramics (LTCC). The concept is to use pre-structured ceramic surfaces with cavities that can be filled with polymeric material locally molten during laser transmission welding. The laser processing used to create appropriate surface textures is described. A variation of the surface pattern was performed and analysed with respect to mechanical bond strength. It was discovered that an increased structure density leads to and increased bond strength reaching a maximum value of 5.2 MPa. Furthermore a technique based on laser transmission welding to replicate surface topographies down to the nanometre scale is described.

1. Introduction

Laser transmission welding is a technology which has attracted considerable interest in recent years [1-5]. It is typically used to join similar thermoplastic materials with high speed and high precision avoiding the use of adhesives [6]. In this process, the moving laser beam passes through the upper transparent material and then penetrates onto the opaque surface of the lower material. Therefore the entire or a major part of thermal energy of the laser beam is absorbed by the opaque part and induces the required heat at the interface between both materials. Subsequently, the opaque part will heat up and the heat will be transferred to the transparent part by thermal conduction which will cause the two materials to be softened at the interface. Finally, a joining area is formed between the two materials.

For certain functionalities however metal or ceramic parts are required, which also require a reliable joining technique. Compared to polymers ceramics are typically significantly more expensive. To maintain low costs, only a minor part, e.g. a sensor or actuator needs to be composed of the more expensive ceramic part while the major part is made of a cheaper polymer. Furthermore, ceramic materials are not transparent and therefore not suited for optical analyses. This functionality can be added by locally inserting a polymeric part.

Laser joining of dissimilar materials has been the subject of intensive studies in recent years [7-9]. Malecha for example demonstrated a bonding technique based on plasma surface modification which enabled a joining of polydimethylsiloxane (PDMS) to low-temperature co-fired ceramics (LTCC) by increased adhesion [10]. A laser fusion technique was proposed by Holtkamp and Olowinsky where a incompatible polymer, metal or ceramic is joined to a transparent polymer by laser transmission welding [11]. This method is based on a macroscopic penetration of the metallic sample into the molten thermoplastic polymer. After re-solidification a mechanical interlock is formed between the two parts by significantly deforming the polymeric material. The strength of such a joint is of course directly correlated to the topography of the surface non-molten material.

Experiments have been performed to successfully use laser transmission welding to join transparent polymers with metals [12, 13]. However, these are based on structuring the metallic part during the welding process and simultaneously melting the polymeric part due to heat conduction. The high temperatures applied during can cause polymer decomposition, bubble formation and long cavity formation at the joint centre which is also related to polymer roasting, charring, vaporization and degradation at the joint interface which may weaken the joint strength and negatively influence the functionality [12].

This study to successfully join ceramic and polymeric is based on prestructuring the ceramic part using a laser process to manufacture defined and reliable surface patterns. The polymeric part will then be locally molten indirectly by heating the ceramic surface using laser transmission welding. The molten polymer will re-solidify inside the laser-generated surface structures to allow a mechanical joining of both parts. Therefore different surface patterns were studied with respect to their capability to allow the joining of ceramics and polymers. As thermoplastic material polymethylmethacrylate (PMMA) was chosen; LTCC was used as ceramic counterpart.

Polymeric materials are widely utilized for durable goods applications due to their low cost, weight to strength ratio, and ease of fabrication. PMMA, especially, has been widely used in architecture and automobile industry [14], but also for biological [15] and micro-analytical systems [16] due to its superior optical, mechanical and chemical properties and low cost.

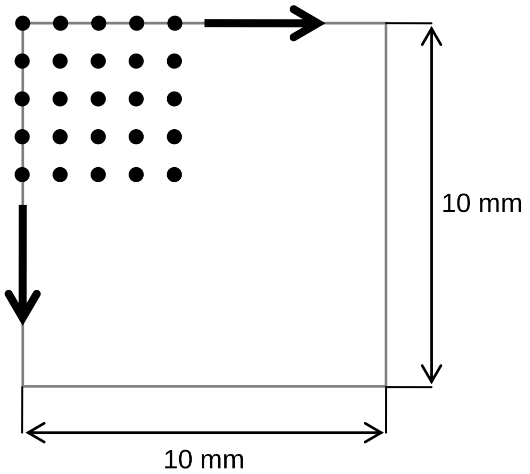
Low temperature co-fired ceramics are materials commonly used in electronics e.g. sensors [17, 18], but also in automotive [19] and microfluidic applications [20-22].

Three types of laser sources have been compared that can be used to machine green ceramic LTCC material by Yung and Zhu – namely excimer (KrF, 248 nm), ultraviolet (Nd:YAG, 355 nm) and infrared solid state (1090 nm) – the infrared laser

has been reported to be the most suitable source for fabricating structures on a green ceramic material [23, 24].
Polyetheretherketone (PEEK) exhibits high chemical and thermal stability, however at considerable material costs. It can efficiently be structured using an excimer laser [25].

2. Experimental

For patterning of the LTCC surface a solid state Nd:YAG laser system has been applied. It operated at a wavelength of 1064 nm using a scanner system with f-theta lens and a focal length of 100 mm. The maximum power of the q-switched laser source was 16 W.
Figure 1 depicts the laser patterning strategy for the LTCC surface. A quadratic array of dimples is manufactured on a surface area of 10 x 10 mm^2. The amount and, correspondingly, the distance between the individual dimples were varied. For this study three different patterns with 17 x 17, 34 x 34 and 50 x 50 dimples were investigated, respectively.



dimples per cm^2	pitch
17 x 17	~0.6 mm
34 x 34	~0.3 mm
50 x 50	~0.2 mm

Figure 1: Schematic of the laser texturing pattern on LTCC surfaces.

For patterning of PEEK an excimer-based process has been applied which has been investigated in a previous study [25]. Laser texturing was performed with an ATLEX-500-SI short pulse excimer laser operating at a wavelength of 248 nm and a maximum repetition rate of 500 Hz. The laser pulse length was 4-6 ns (FWHM). The ATLEX short pulse excimer laser generated a raw “flat-top” beam directly applicable without homogenizing devices. The object lenses used had a demagnification factor of 1:10 and enable maximum laser fluences of 5 J/cm^2

which can be attained on sample surfaces. A high precision x-y-z position system enables a positioning accuracy better than 500 nm.
For laser transmission welding a high power diode laser (FLS IronScan, Fisba Optik AG) with a maximum laser output power of 50 W and a wavelength of 940 nm was used. The laser beam is focused onto the sample surface by an objective lens (f-theta objective, focal length 163 mm) with a laser focus diameter of about 1 mm. Via deflection mirrors the beam is scanned over the sample surface at speeds up to 2000 mm/s. This enables the treatment of an area of 100x100 mm^2 without using a positioning stage for the samples. Furthermore, the use of a galvo-scanning mirror system avoids significant changes in the feed rate during processing. The temperature in the heat-affected-zone is measured on-line during the annealing process with a pyrometer (FLS PyroS, Fisba Optik AG), that directly controls the laser power. If the temperature increases, e.g., because of dust particles or layer inhomogeneity the laser power is adjusted within several milliseconds. The control interval of the monochromatic pyrometer system is about 1 ms. The process temperature can be controlled in the range of 120°C up to 700°.
The samples were set on a pneumatic stage and then pressed against a transparent fused silica plate to ensure a direct contact of the welded parts. All laser processes were performed in ambient air.
The mechanical testing of the bond strength was performed using a tensile strength testing setup (UTS-Universalprüfmaschine, Zwick-Roell). The welded samples were stressed until failure while force and strain were measured. During the mechanical measurements the test speed was maintained at 25 mm per minute. The bond strength s was determined using the structured sample area of $A = 10 \times 10$ mm^2 and the maximum force F_{max}:

$$s = F_{max} / A \quad (1)$$

3. Results and Discussion

Three different textures were used to investigate the bonding properties of PMMA and LTCC. Using laser transmission welding the polymeric part is designed to be melted on the surface and flow into the structures created on the ceramic surface. The driving force for the complete filling of the cavities is the capillary force resulting from the overall reduction of the surface energy.
By directing the laser beam over the surface in a circular motion, round dimples were created on the LTCC surface. A laser scanning speed of 50 mm/s with an average power of 2.74 W at 5 kHz were used to create the cavities sown in Figure 2. The diameter of the structures was about 100 µm.
It can be seen in the cross-sectional image of a dimple that the surface of the laser-generated structure shows a high roughness. For this application this is advantagous since it allows a mechnical interlocking of the two materials. In

addition an increased surface area leads to increased bonding surface. Smooth ablation of LTCC is possible by adjusting the laser parameters [23, 24]. This is, howerever, not desireable for this application.
For laser transmission welding a welding temperature of 160°C was selected, as higher welding temperatures resulted in bubble formation in the PMMA part. Lower tempertures typically lead to decreased bond strenght [16].

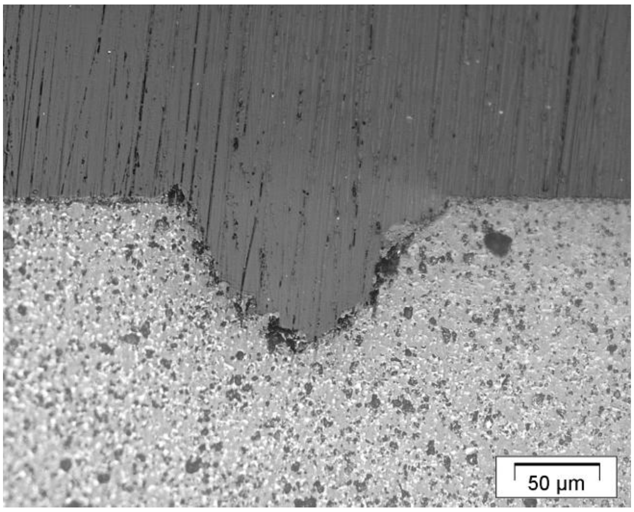


Figure 2: Cross-section image of laser structured dimple in LTCC.

Bonding of unstructured LTCC samples with PMMA was not possible as insufficient adhesion was achieved. Therefore, a surface modification such as laser patterning is required to connect the two materials. The bond strengths of the samples with the three investigated surface patterns joined using laser transmission welding are shown in Figure 3.
The surface patterns exhibit a significant influence on the bond strength. The pattern with the lowest dimple density showed the lowest bond strength of 1.8 MPa. By increasing the structure density an increase of bond strength to 4.7 MPa and 5.2 MPa was measured for the 34x34 and 50x50 dimple samples, respectively.
The direct correlation between structure density and bond strength can be easily explained by the increased surface area and increased mechanical interlocking due to increased amount of dimples per area.
Compared to the tensile strength of bulk PMMA of ~70 MPa [26] the bond strength obtained in this study is relatively low. However, Sun et al. measured a bond strength of 2.15 MPa for thermal bonding of PMMA [27]. Compared to this value the bond strength could be increased more than twofold by pre-structuring the LTCC surface. The plasma-based method described by Malecha exhibited bond strengths of about 0.26 MPa [10].

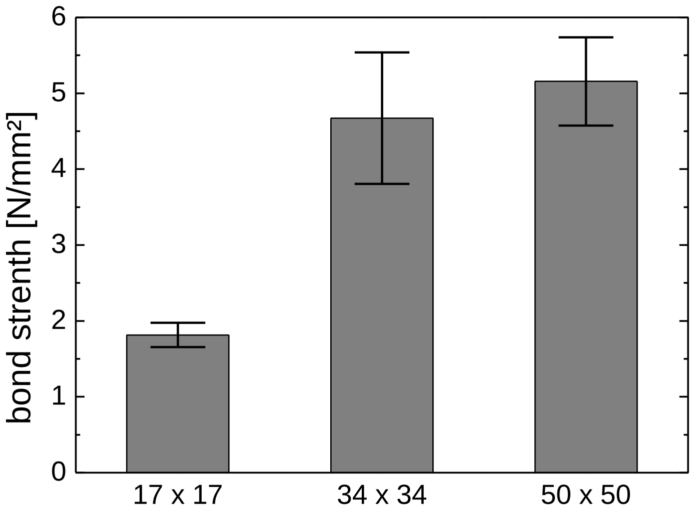


Figure 3: Bond strength for different structuring designs as function of dimple amount per 1 cm^2.

Further proof of the improved bonding properties can be obtained by investigating the fractured parts after mechanical testing. Figure 4 shows examples of fractured samples after tensile strength testing. The joint of the sample with the lowest structure density of 17 x 17 clearly failed at the welding interface between the PMMA and LTCC. No ceramic residues were visible on the PMMA surface. In contrast, the samples with higher structure density of 34 x 34 and 50 x 50 failed in the ceramic component rather than the polymeric part. This is an indication of the improved joint strength and correlates well with the bond strengths obtained by mechanical testing. Cracks formed during laser patterning could be responsible for the low mechanical stability of ceramic material.

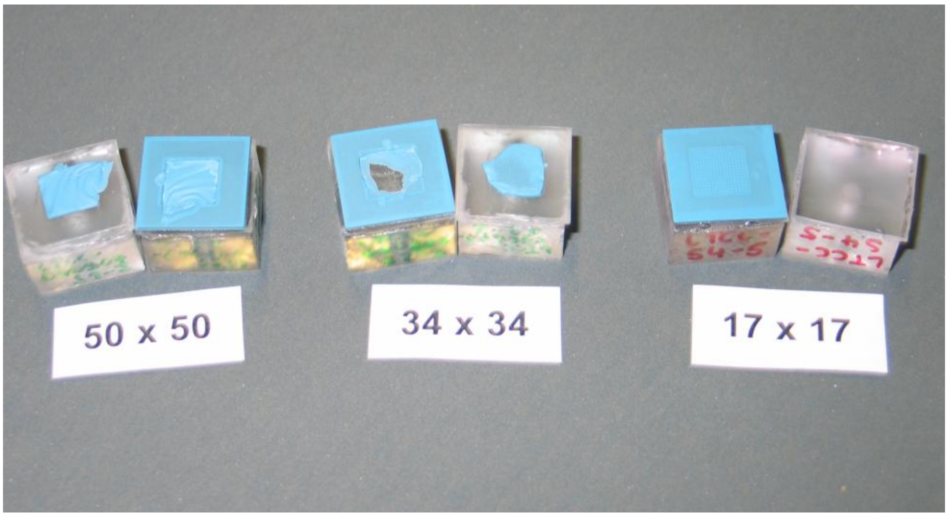


Figure 4: Samples of PMMA welded to laser structured LTCC with three different textures after tensile for different number of dimples per 1 cm^2.

During close investigation of the 17 x 17 sample it was discovered that the polymeric structures penetrating the ceramic cavities were not broken by mechanical testing but rather demoulded from the weld joint. Therefore a negative replica of the laser-generated dimples and the surrounding surface topography on the polymeric part could be obtained and is displayed in Figure 5 (right). It can be seen that the structures LTCC have been filled completely with molten PMMA during the laser welding process. This behaviour is not self-evident as the parts are pressed together before and during laser transmission welding which possibly could entrap air inside the cavities of the ceramic part.
However, this can be attributed to the process strategy used in laser transmission welding. The laser is scanned laterally over the sample surface and therefore the surfaces are welded not simultaneously but sequentially. The air can consequently escape and is not trapped inside the laser-generated dimples.

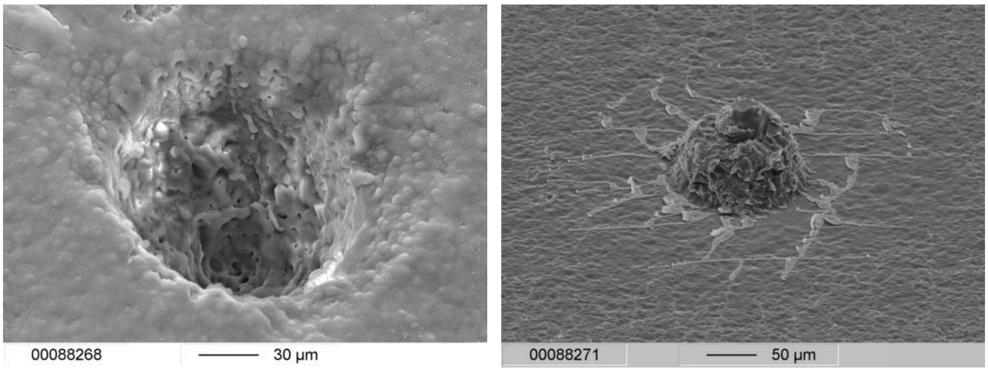


Figure 5: SEM images of the LTCC (left) and PMMA (right) surfaces after mechnical testing.

As a result it could be demonstrated, that laser transmission welding is as processing technique can be applied to mould transparent polymers.

To further investigate this innovative process, channel structures were created in a high-temperature resistant polymer, PEEK, via excimer laser structuring. The channel with a width and depth of 50 µm was manufactured using mask imaging.

The positive and negative parts i.e. PEEK and PMMA, manufactured by laser structuring and laser transmission welding with subsequent demoulding, respectively, are depicted in Figure 6. Remarkably, the replication has a very high precision, reproducing the surface topography down to the nanometer scale.

The demoulding of the parts is possible by using force if the structure density is not too high or the structures have a smooth surface. A drop of organic solvent (e.g. isopropanol or ethanol) can be used to ease demoulding without damaging the polymer.

An important advantage of this technique is, that only localized heating of the sample occurs which results in (i) fast processing times as heating and the cooling times are short, (ii) low thermal impact on the polymeric sample e.g. microstructures not in the direct vicinity of the joining surface remain intact.

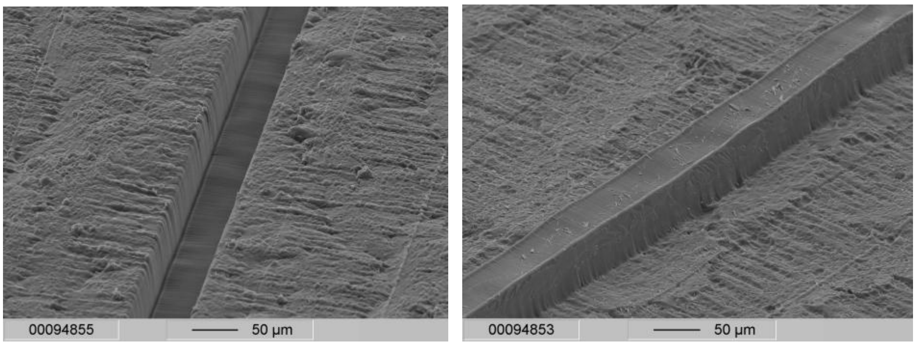


Figure 6: SEM images of laser structured PEEK (left) and the PMMA counterpart after laser transmission welding (right).

As final experiment microstructures in silicon were used as mould. Replication of these structures onto PMMA could be successfully demonstrated. Figure 7 depicts the two counterparts. Reproduced roughness on the nanometer scale is clearly visible on both surfaces.

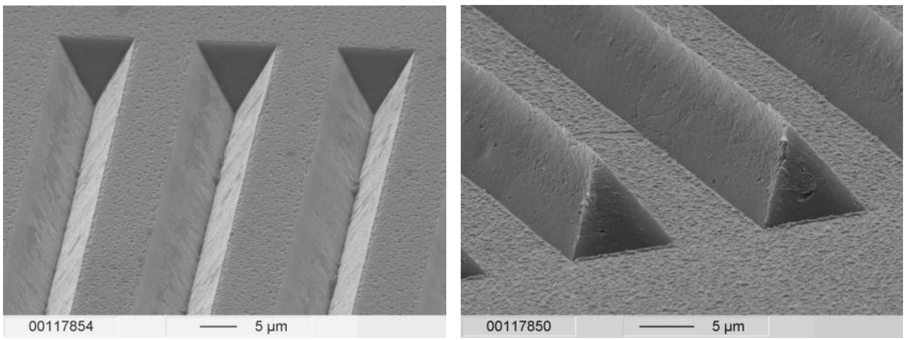


Figure 7: SEM images of microstructures in silicon (left) and a PMMA sample demoulded after laser transmission welding.

4. Summary

A method for joining a polymeric (PMMA) and a ceramic material (LTCC) based on laser transmission welding and pre-structuring has been investigated. Therefore dimples with a diameter of 100 µm and high surface roughness were manufactured on the LTCC surface. Via heating the LTCC surface through the transparent polymer the PMMA is melted locally and created a mechanical bond between both parts. The pattern on the LTCC surface was varied and higher structure densities resulted in higher bond strengths.

The joining method here proposed can be adapted for joining not only thermoplastic polymer and ceramics with but also to duromers or metals.

Furthermore a laser assisted moulding technique is proposed by which is able to mould thermoplastic polymers by local laser transmission welding.
During mechanical investigation it was found, that laser transmission processing can also be used to replicate surfaces onto transparent polymers nanometer scale.

Acknowledgement

We would like to express our thanks to M. Beiser for his assistance in laser processing and N. Kandora for support in SEM imaging. We would like to acknowledge the financial support from the European Commission within the INTERREG NWE IVB programme for the project ECOLASERFACT (project number 268l). Finally, the support by the Karlsruhe Nano Micro Facility (KNMF) for laser processing is gratefully acknowledged.

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