Available online at www.sciencedirect.com
ScienceDirect
Journal of the European Ceramic Society 29 (2009) 899–904
www.elsevier.com/locate/jeurceramsoc

Manufacture of 3D structures by cold low pressure lamination of ceramic green tapes

Karin Schindler, Andreas Roosen *

University of Erlangen-Nuremberg, Department of Materials Science, Glass and Ceramics, Martensstraße 5, 91058 Erlangen, Germany

Available online 30 December 2008

Abstract

3D multilayer devices were generated by Laminated Object Manufacturing (LOM), a well-known rapid-prototyping technology. Divergent from this method, commercial ceramic green tapes were used which were laminated by Cold Low Pressure Lamination (CLPL). In contrast to thermo-compression, which works at pressures and elevated temperatures, CLPL allows to join particularly fine, complex structures with cavities or undercuts, because no mass flow occurs. This technique is based on gluing the adjacent tapes by means of an adhesive film at room temperature under a low pressure. After binder burnout and sintering the ceramic laminate has a homogenous and dense microstructure free of interfaces. This modified LOM technique is particularly suitable for the production of Micro Electro Mechanical Systems (MEMS).

In the given paper commercial Low Temperature Co-fired Ceramic (LTCC) green tapes were used, which were structured by means of a high-frequency milling plotter and laminated by using CLPL. Various 3D devices of different shape with inlying cavities were manufactured. The quality of the fired and unfired structures of the devices were characterised by different methods and show a high quality surface of the multilayer structures. Process aspects of the CLPL technique are discussed. The results demonstrate the advantages of this method for the fabrication of MEMS.

Keywords: Shaping; Joining; Lamination; Glass ceramics; Functional applications

1. Introduction

Laminated Object Manufacturing (LOM) is an established rapid-prototyping technique. The high building speed and low operation costs of this manufacturing process are of interest for many applications.[1] LOM was originally developed to form 3D paper models to test new designs or functions.[1,2] Paper had the advantage of a cheap material.[1] In the LOM technique the prototype is constructed layer-wise by building up the given sequence of the sliced CAD model. The contours of every sheet are cut by a laser or knife. The grid pattern is also cut in the unused part of the layer to form cubes which can be removed relatively easy after the object is completed. During the LOM process this excessive material is left in the building block as a support structure for the next sheet.[1] To join the layers the bottom side of the paper is coated by a heat-sensitive adhesive. The adhesive melts during applying temperature by a hot lamination roller.[3] The procedures of stacking, bonding and cutting are repeated until the complex solid is formed. Finally, the excessive material must be manually removed from the part, which is called de-cubing. This process step is time-consuming and labour-intensive depending on the geometrical complexity of the parts.

Meanwhile, not only paper models are made by LOM. Functional devices of various materials are of interest in many application fields. Metal parts, e.g., offer high toughness and strength.[4,5] However, metal sheets are more difficult to fabricate by LOM due to higher sheet-thickness up to 1 mm.[4] Compared with the sheet-thickness of paper which is about 50–100 μm, respectively, the model accuracy of thicker metal sheets is lower.[4] To improve the surface quality the steps which are formed by the different tape layers must be removed by finish processing.[4] The metal sheets were bonded by coating the single layers with an alloy of a low melting point.[4,5]

Ceramic parts made by LOM are used, e.g., for medical applications like wrists, hips or dental implants.[6] The ceramic models were directly created by scanning the human parts by computer tomography. Green tapes of alumina, zirconia, hydroxyapatite or calcium phosphate were used.[6] The process of lamination is not described. Klosterman et al. produced monolithic ceramics as well as ceramic matrix composites for structural applications by LOM.[7] Ceramic green tapes were laminated by using a heated roller which applies temperature and pressure to join the layers.[7]

* Corresponding author. Tel.: +49 9131 8527547; fax: +49 9131 8528311.
E-mail address: Andreas.Roosen@ww.uni-erlangen.de (A. Roosen).

0955-2219/$ – see front matter © 2008 Published by Elsevier Ltd.
doi:10.1016/j.jeurceramsoc.2008.07.041

The temperature has to be in the range of 120–180 °C to melt the binder and to cause inter-layer adhesion. However, thermal gradients caused the formation of cracks or warpage and the tapes tend to stick at the lamination roller if it is heated above the softening point of the binder system.[7] To overcome these problems a solvent was sprayed onto the layers to reduce the lamination temperature. But this method results in increased difficulties of the de-cubing process. In addition, a final post pressing step between heated plates at a low pressure for 60 min is necessary to achieve a laminate of sufficient quality for further processing.[7]

In general, ceramic green tapes were laminated via thermo-compression by applying temperature and pressure. To join, e.g., Low temperature Co-fired Ceramic green tapes by this technique, temperatures in the range of 60–80 °C and pressures of 10–30 MPa must be applied. These temperatures are above the glass transition temperature of the green tape's binder system. Thereby, the binder system softens and the tapes are joined by the interpenetration of particles from each tape at the interface of two adjacent tapes. However, the main disadvantage of thermo-compression is the occurring mass flow, which destroys fine complex structures or cavities.

To overcome these problems a new lamination technique called Cold Low Pressure Lamination (CLPL) was developed. CLPL works with double-sided adhesive tapes which are positioned between the ceramic tapes.[8] The softening point of these adhesives is below room temperature so that the tapes stick together at room temperature, after applying a low pressure. During binder burnout, the adhesive melts, which results in capillary forces, keeping the individual layers together. After sintering, the tapes are joined. Compared to thermo-compression CLPL avoids mass flow and inhomogeneous densification in case of inlying cavities or fine structured designs. Therefore, this lamination technique is particularly suitable for production of MEMS (Micro Electro Mechanical Systems).

In the present paper, CLPL was used as the lamination technique for the manufacture of 3D structures by the LOM processes. As green tapes, commercial LTCC (Low Temperature Co-fired Ceramics) tapes were used.

2. Experimental procedure

2.1. Raw materials

To produce 3D devices by CLPL, a commercial LTCC green tape of 250 μm thickness was used (DuPont Green Tape 951AX, UK). This tape is a glass-ceramic composite, which is composed of a SiO_2–Al_2O_3–RO glass and Al_2O_3 as a ceramic filler. After firing at 870 °C, the dense LTCC material exhibits suitable properties (Table 1). For joining a double-sided adhesive tape was chosen with a thickness of 12 μm.

Table 1
Characteristic data of Green Tape DuPont 951 AX.

Thickness	250 μm
Green density	2.15 g/cm^3
Sintered density	3.08 g/cm^3
Linear shrinkage in x and y-plane	13%
Linear shrinkage in height	20%
Mechanical strength[a]	320 MPa
Thermal conductivity[a]	3.0 W/m K
Coefficient of thermal expansion[a]	5.8×10^{-6} K^{-1}

[a] Data of manufacturers,[9] fired state.

2.2. Manufacturing process

The green tapes were structured by means of a milling plotter ProtoMat® C100/HF with a high frequency spindle (LPKF Laser & Electronics, Garbsen, Germany) (Table 2). The rotational speed of the frequency spindle and its motion in x–y direction is variable. The motion of the spindle in z-direction is continuously adjustable by a micrometer screw. Different tools for milling, drilling and cutting can be used for machining. During machining the green tapes were fixed by means of a self-adhesive pad. Table 2 summarizes the most important data of the milling plotter. The design, which has to be generated by the plotter, was developed by CAD software, followed by the transformation into Gerber data.

The structured tapes were laminated by CLPL at low pressures and ambient temperature. The adhesive tape was positioned on top of the green tape with a soft roller. After removing the upper release tape the next green tape was applied. It is important to avoid the formation of bubbles between green tape and adhesive layer during CLPL. Binder burnout and sintering were carried out in a chamber furnace (Nabertherm GmbH, Bremen, Germany) in air. The peak temperature of the sintering profile was 870 °C.

2.3. Characterisation of the green and sintered laminates

Thermo-gravimetric measurements of the green tape, the pure adhesive tape, the adhesive tape in contact with a de-bindered ceramic tape (heated up to a temperature of 500 °C) and of CLP-laminates were performed (DuPont Instruments, USA). The measurements were done in air up to temperatures of 600 °C with a heating rate of 1 K/min.

The density of the green tape was characterised by measuring sample size, thickness and weight. The sintered densities were measured by buoyancy (AG 204, Mettler Toledo, USA). The linear shrinkage was determined by geometrical measurements before and after firing.

To characterise the densification behaviour of a two-layered CLP-laminate, samples were heat treated at different temperatures between 100 and 500 °C and quenched in cold air. The samples were cut and polished and analysed with the Scanning Electron Microscope (SEM, Quanta 200 FEI, Netherlands).

Table 2
Equipment data of milling plotter ProtoMat® C100/HF (data of manufacturer).

Rotational speed	10,000–100,000 rpm
Traversing range	340 mm × 200 mm
Resolution (smallest step)	7.94 μm
Repeat accuracy	±5 μm
Drilling performance	Up to 120 swings/min
Speed of movement	Up to 40 mm/s

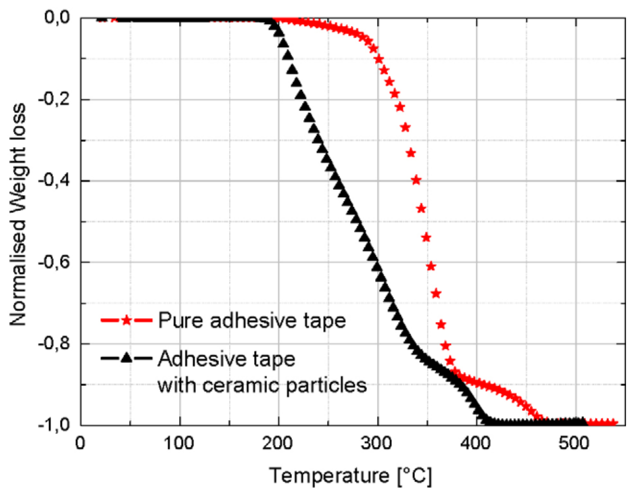

Fig. 1. Thermal decomposition of the adhesive tape.

The wetting behaviour of the melting adhesive on the ceramic green tape was analysed by means of an optical dilatometer.[10] For the investigation, a piece of adhesive was positioned on the green tape and both were heated up using the normal sintering profile.

Sintered laminates were analysed by taking micrographs of the cut and polished cross sections with an optical microscope (Stereomicroscope Leica M420, Leica Microsystems, Germany) and a Scanning Electron Microscope, respectively.

3. Results and discussion

3.1. Thermo-gravimetric measurements

Cold Low Pressure Lamination requires knowledge of the decomposition behaviour of the adhesive and of the organic components of the green tape.

Fig. 1 shows the result of the decomposition behaviour of the pure adhesive film and of the same adhesive film, but in

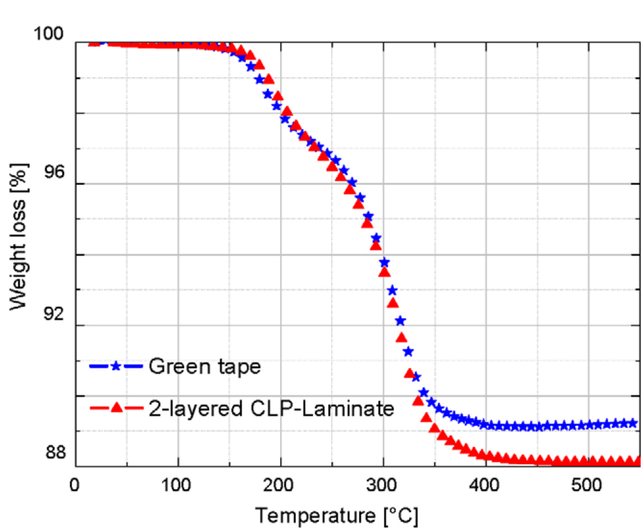

Fig. 2. Thermal decomposition of the green tape and a two-layered CLP-laminate.

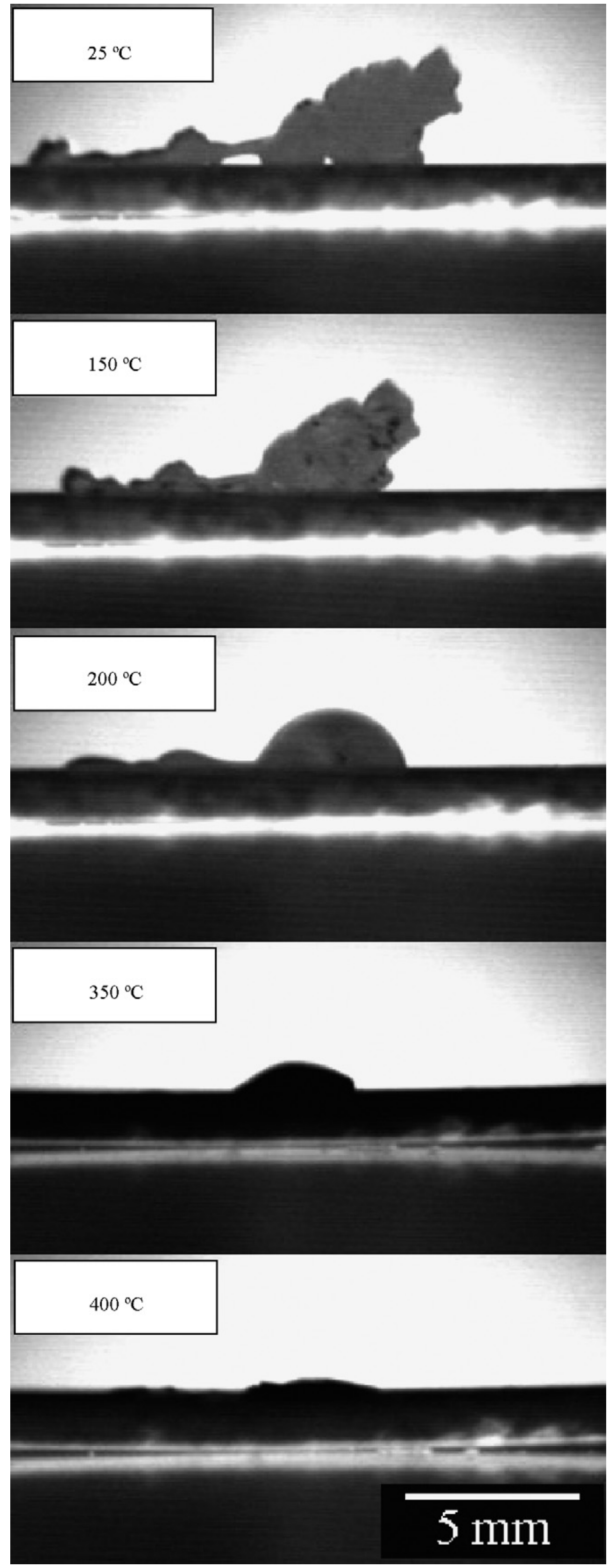

Fig. 3. Wetting behaviour of the adhesive during heating.

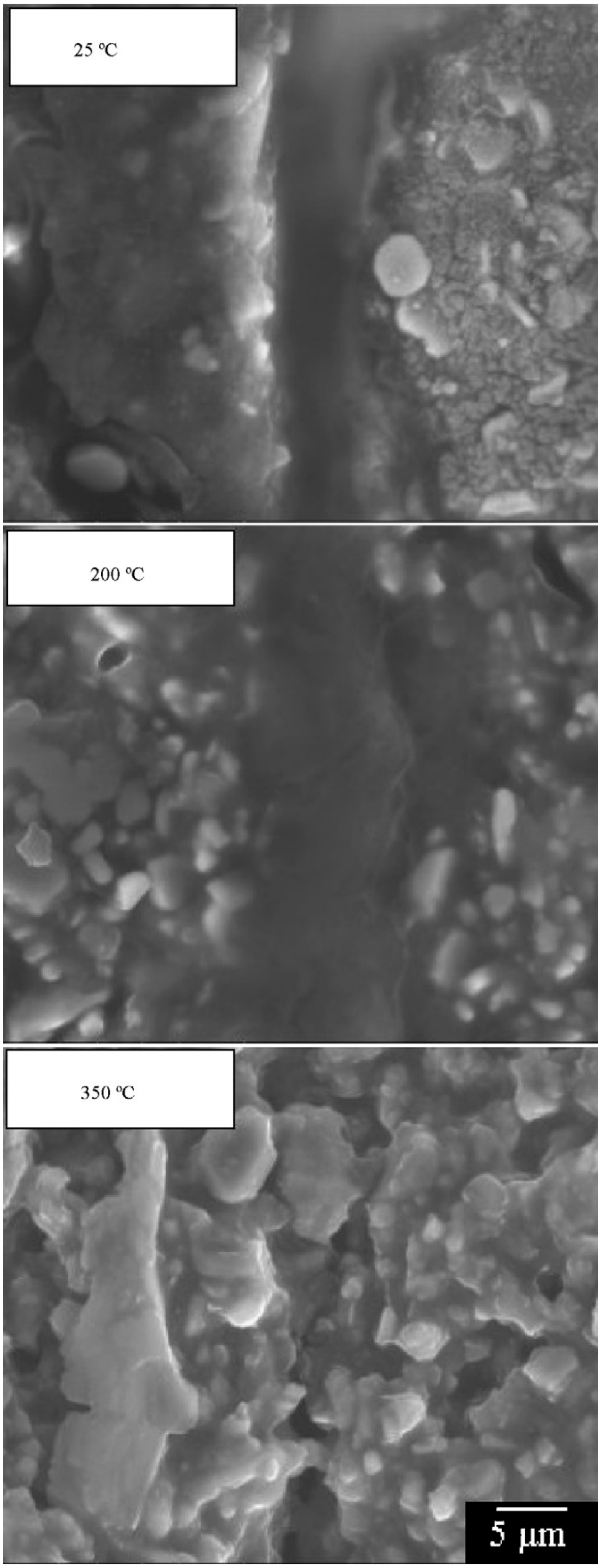

Fig. 4. SEM micrographs of a polished cross section of a two-layered CLP-laminate removed at different temperatures from the kiln.

contact with a de-bindered ceramic tape. The decomposition of the adhesive starts slowly at 200 °C and is finished at 460 °C. The catalytic effect of ceramic particles on decomposition of the adhesive shifts the curve to lower temperatures. In this case the decomposition of the adhesive takes place much faster and is already finished at 410 °C. This knowledge is important for the determination of the temperature profile during binder burnout, when the final joining process takes place.

At the same time when the adhesive melts, the binder burnout of the green tapes takes place. In Fig. 2 the thermo-gravimetric analyses of the used LTCC green tape and a two-layered CLP-laminate are shown. The two-layered laminate with the adhesive layer exhibits a weight loss of 12 mass%, which is 1 mass% higher than for the single green tape, due to the additional adhesive tape. The decomposition of the CLP-laminate starts at 150 °C, which is caused by the decomposition of the binder system of the green tape. The decomposition is finished at 425 °C. The results show that the decomposition of the binder system of the green tape leaves a porous green tape, which can be infiltrated by the liquid polymer melt of the adhesive. This results in capillary forces, which holds together the individual tapes.

3.2. Characterisation of the CLPL process

The behaviour of the molten adhesive on the green tape is shown in Fig. 3. The photos, which were taken by the optical dilatometer, show that the adhesive becomes liquid around 200 °C. At this temperature it wets the surface of the ceramic tape, but the viscosity is relatively high. During further heating the viscosity decreases. Partially it infiltrates the porous ceramic tape.

The changes at the interface of a CLP-laminate with its adhesive film are demonstrated in Fig. 4. The SEM micrographs were taken at room temperature after the removal of the specimen from the kiln at the given temperatures. The adhesive film can be clearly seen in the green state and at 200 °C

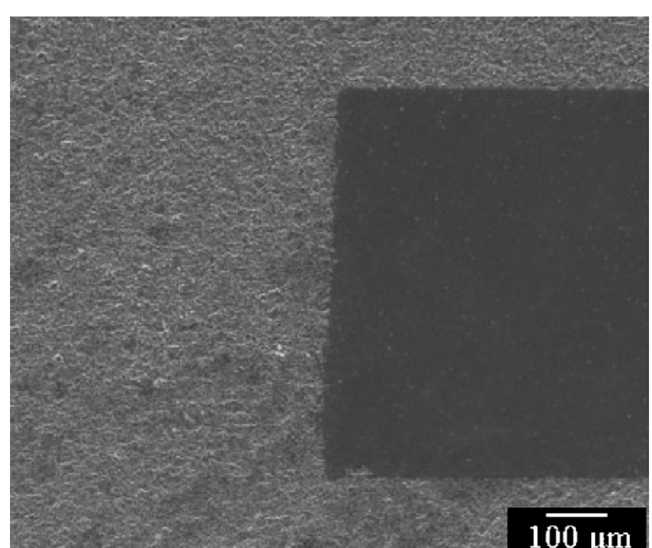

Fig. 5. SEM micrograph of a polished cross section of a sintered 10-layered CLP-laminate with an inlying cavity (dark area).

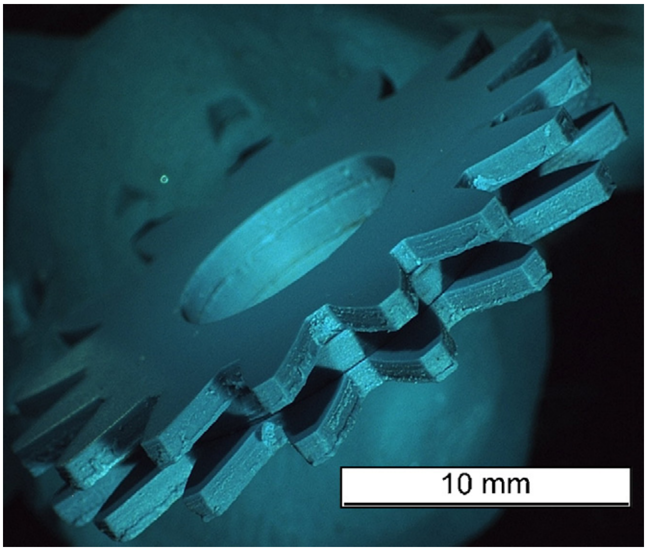

Fig. 6. Gear wheel with a groove made by CLPL of 15 LTCC green tapes.

when the adhesive was already molten. At 350 °C when the decomposition of the adhesive is nearly finished, almost no interface between the ceramic tapes can be detected. After sintering the CLP-laminate is dense with densities of about 99% TD (theoretical density) and no interfaces can be seen anymore (Fig. 5).

The main advantage of CLPL compared with the thermo-compression technique of the current LOM process is the possibility to join the green tapes at room temperature using only a slight pressure. This simplifies the LOM process for ceramic green tapes, because both can be used from a roll. A final post-pressing treatment of the whole stack is not necessary. With the possibility of mechanically milling the final devices can be cut out of the whole laminate with its inlying cavities. The obtained multilayer device exhibits surfaces of high quality without steps from the individual tape layers.

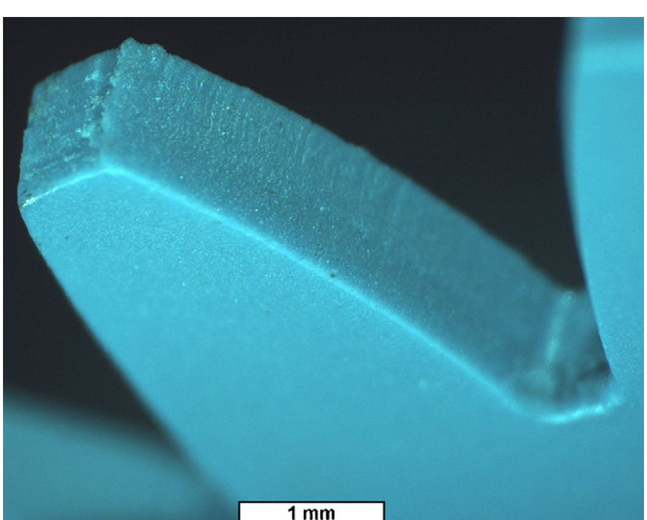

Fig. 7. Magnification of a part of the gear wheel.

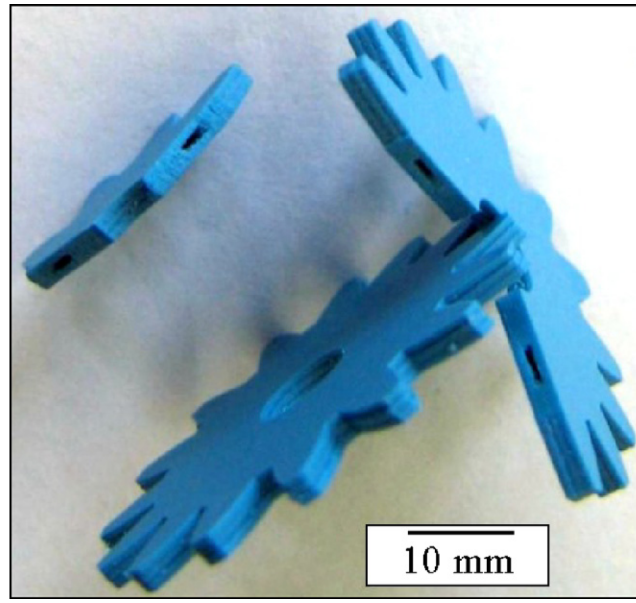

Fig. 8. Complex structured gear wheels with an inlying cavity made by CLPL of 6 LTCC green tapes.

3.3. *Characterisation of multilayer devices*

CLPL is an advantageous lamination technique for the manufacture of complex 3D devices. Figs. 6–8 demonstrate the design possibilities of this technique. Fig. 6 shows a gear wheel with a groove made from 15 LTCC green sheets via CLPL. Fig. 7 shows a magnification of the surface of such a gear wheel and illustrates the high model accuracy and surface quality of this technique. In Fig. 8 there are two gear wheels with an inlying cavity. All parts are fully densified.

These results show, that the application of CLP-lamination in the LOM process allows to form 3D devices of any outer shape as well as internal structure. The method has the advantage to offer high model accuracy combined with high building speed.

4. Conclusion

The paper describes the combination of the lamination technique Cold Low Pressure Lamination with the LOM technique. This lamination technique is of advantage for fast fabrication of multilayer devices of fine and complex structures with inlying cavities or channels like it is required in MEMS. The advantage of CLPL is the almost pressure-less lamination process which can be performed from the roll without applying temperature. By the use of commercial LTCC green tapes, high quality, dense multilayer devices can be manufactured at low costs.

References

1. Cho, I., Lee, K., Choi, W. and Song, Y.-A., Development of a new sheet deposition type rapid prototyping system. *Int. J. Mach. Tools Manuf.*, 2000, **40**, 1813–1829.
2. Chiu, Y. Y., Liao, Y. S. and Hou, C. C., Automatic fabrication for bridged laminated object manufacturing (LOM) process. *J. Mater. Process. Technol.*, 2003, **140**, 179–184.

3. Pham, D. T. and Gault, R. S., A comparison of rapid prototyping technologies. *Int. J. Mach. Tools Manuf.*, 1998, **38**, 1257–1287.
4. Yi, S., Liu, F., Zhang, J. and Xiong, S., Study of the key technologies of LOM for functional metal parts. *J. Mater. Process. Technol.*, 2004, **150**, 175–181.
5. Obikawa, T., Yoshino, M. and Shinozuka, J., Sheet steel lamination for rapid manufacturing. *J. Mater. Process. Technol.*, 1999, **89–90**, 171–176.
6. Griffin, A., McMillin S., Griffin, C., and Barton, K., Bioceramic RP Materials for Medical Models. In: Proceedings of the 7th International Conference on Rapid Prototyping, 1997, pp. 355–359.
7. Klosterman, D., Chartoff, R., Osborne, N., Graves, G., Lightman, A. and Han, G., Laminated Object Manufacturing (LOM) of advanced ceramics and composites. In *Proceedings of the 7th International Conference on Rapid Prototyping*, 1997, pp. 43–50.
8. Piwonski, M. and Roosen, A., Low pressure lamination of ceramic green tapes by gluing at room temperature. *J. Eur. Ceram. Soc.*, 1999, **19**, 263–270.
9. DuPont Microcircuit Materials, 951 Green Tape^{TM}. Product Data Sheet, 2001.
10. Wagner, M., Roosen, A., Stiegelschmitt, A., Schwanke, D. and Bechthold, F., In-situ shrinkage measurements of LTCC multilayers by means of an optical dilatometer. In *European Ceramics VII*. Trans Tech Publishers, Switzerland, 2002, pp. 1218–1284.