

# Flexible and Printed Electronics



## PAPER

# Inkjet-printed low temperature co-fired ceramics: process development for customized LTCC

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## Abstract

This paper investigates the utilization of digital printing technologies for the fabrication of low temperature co-fired ceramics (LTCC). LTCC offer great opportunities for applications such as antennas, sensors or actuators due to their outstanding properties like low dielectric loss, low permittivity, low coefficient of thermal expansion and at the same time high reliability in harsh environments (heat, humidity, and radiation). LTCC are multilayer circuits that are typically functionalized by screen-printing. This publication investigates the replacement of screen-printing by digital printing processes, such as inkjet and Aerosol Jet printing, to facilitate a more resource-friendly and customizable manufacturing of LTCC. The use of digital printing technologies not only streamlines small-scale productions and development processes but also offers the advantage of achieving miniaturization down to single-digit microns. In this publication, digital printing processes, filling of vias, lamination processes, co-firing at 850 °C and printing on fired LTCC were investigated. Three layers of nanoparticle silver ink were printed on green LTCC tape and 100% of the embedded printed structures were conductive after co-firing. Filling of vias with inkjet printing was investigated and the most important process parameters were found to be the clustering of vias, the amount of active nozzles and the substrate temperature. Printing on fired LTCC demonstrated high precision, and sintering at 600 °C achieved strong adhesion of printed structures to LTCC. These successful findings culminate in presenting a process chain for fully maskless structured, multilayer LTCC.

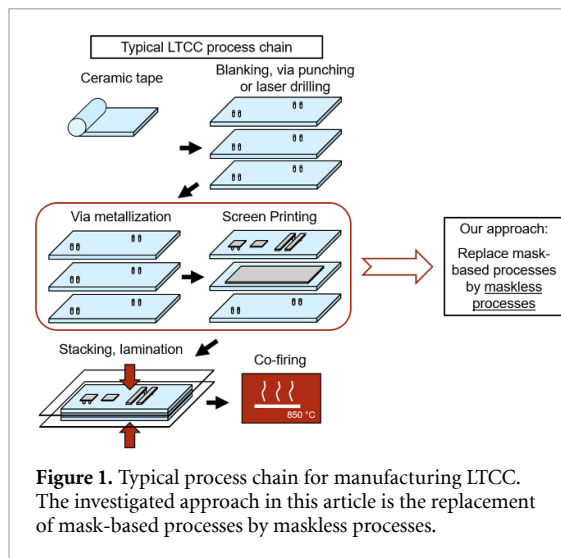
## 1. Introduction

Multilayer ceramic circuits based on low-temperature co-fired ceramics (LTCC) have been used in automotive technology, telecommunications and medical technology for around 25 yrs. LTCC are multilayer glass ceramic substrates, sometimes referred to as glass ceramics, because their main composition typically consists of glass and alumina as a filler [1]. The LTCC technology is characterized by a high degree of miniaturization and at the same time the highest level of reliability. The properties of LTCC include the mechanical rigidity and hermeticity, which are important for applications that require highly reliable and inert environment [2]. Furthermore, the low-loss behavior when used at high frequencies is worth

mentioning, which makes the technology suitable for millimeter wave applications [3].

The layers of LTCC can be individually structured and functionalized. Cavities and channels can be integrated using punching or laser ablation, and each level is provided with conductor tracks, through-contacts (so-called vias) or passive components, such as resistors, coils, capacities.

LTCC are typically functionalized by screen printing. However, for small batch sizes or research and development, this technology has some disadvantages, such as costly and time-consuming manufacturing of screens. Digital printing technologies have the advantage that fast layout changes can be performed without the need for screens or other masks. This saves money and time, and provides the ability



**Figure 1.** Typical process chain for manufacturing LTCC. The investigated approach in this article is the replacement of mask-based processes by maskless processes.

to manufacture economically down to batch size 1. Especially for those reasons, this paper investigates the manufacturing of LTCC by maskless technologies, like inkjet or Aerosol Jet printing. For filling vias, a maskless squeegee process is investigated as well. Figure 1 illustrates the typical process chain for manufacturing LTCC and points out the process steps, investigated in this paper. First, vias are laser drilled, then metallized and conductive structures are printed on the ceramic tape. After the printing processes, the sheets are stacked, laminated and co-fired. The literature shows that structuring LTCC with digital printing processes can be possible [4, 5]. An approach of manufacturing LTCC solely by maskless processes, has however not been presented to the knowledge of the authors.

Digital printing technologies include multiple approaches, such as inkjet printing, Aerosol Jet printing, dispensing, micro dispensing, electrohydrodynamic printing, laser induced forward transfer and piezo jetting. For those technologies, print jobs are based on digital information, like pixel-based files, G-Code or CAD-CAM based toolpaths. This provides the benefit that ink is only applied, where it is intended to, which saves important resources.

Inkjet printing is known to achieve a high throughput of up to  $200 \text{ m min}^{-1}$  printing velocity and is well qualified for sheet based processes, such as metallizing LTCC sheets [6]. Such high throughputs can be achieved by high drop spacings, high frequencies or print heads with large droplet sizes. For printed electronics, the throughput is typically one order of magnitude lower. For finer lines and narrower drop spacing, the throughput decreases, and a compromise between fine line width and high throughput has to be made. Drop sizes of  $15 \mu\text{m}$  can be achieved with 1 pL print heads [7]. Low layer heights of  $0.5\text{--}5 \mu\text{m}$  are typically achieved with almost zero waste due to the drop on demand processes. For

even narrower layouts, femtoliter print heads or other digital printing technologies like Aerosol Jet, microdispensing or electrohydrodynamic printing can be utilized. Depending on the printing technology, line widths of around  $1 \mu\text{m}$  can be achieved [8].

Structuring LTCC by digital printing technologies opens up a lot of important research areas. Those include developing the printing process itself, contacting different layers within the LTCC through vias, lamination, co-firing at  $850 \text{ }^\circ\text{C}$  and printing on the sintered LTCC. This paper will investigate those processes.

First, printing processes on LTCC need to be selected and developed. Selecting a suitable ink, adjusting parameters for sufficient wetting and characterizing the printed structures are typical process steps for this development. For structuring LTCC green tapes, this paper investigates inkjet printing. For fine pitch printing on fired LTCC, this paper utilizes Aerosol Jet printing.

After printing on LTCC tapes, filling of vias is investigated. Conventional filling of vias in LTCC-technology is performed with highly viscous pastes. Filling of vias by screen printing was investigated by Kujala [9]. Kujala found that as the via size increased above  $150 \mu\text{m}$ , the vias were not completely filled, and the sidewalls were coated with ink. Furthermore that work shows that ink with higher viscosity of  $20\text{--}30 \text{ Pa}\cdot\text{s}$  fills the vias more effectively than a less viscous ink with  $10\text{--}20 \text{ Pa}\cdot\text{s}$ . In contrast to such high viscosity inks used for screen printing, inkjet printing is usually performed with inks with viscosities of around  $4\text{--}20 \text{ mPa}\cdot\text{s}$ . With such low viscosities, filling of vias faces challenges and needs to be investigated thoroughly. Various publications investigated filling of vias by inkjet printing up to this point. Cummins filled through silicon vias (TSV) with diameters of  $190 \mu\text{m}$  with copper nanoparticle ink, Eiroma filled TSV with diameters of  $45 \mu\text{m}$  with silver nanoparticle ink and Khorramdel filled TSV with diameters of  $80$  and  $85 \mu\text{m}$  vias with silver nanoparticle ink [10–13]. None of those vias were filled completely, but the sidewalls of the vias were covered by ink. They all showed electrical conductivity to achieve vertical electrical interconnections. This proves that filling of vias by inkjet printing can generally work. The sidewall coverage with ink was documented to be mostly good. The most challenging part showed to be the coverage of the upper edge of the vias. Different printing approaches like amount of drops, time between drops, time between layers or substrate temperature seem to influence the wetting [10, 13]. By controlling those parameters, the upper edges of vias may be wetted sufficiently. This paper will investigate vias with small diameters, down to  $40 \mu\text{m}$ . Based on the literature, filling of such vias can be possible, even with low viscous inks. Filling vias on LTCC by inkjet printing and co-firing at  $850 \text{ }^\circ\text{C}$  have not been investigated

and published to the knowledge of the authors. This publication investigates those issues.

After printing, the nanoparticles have to be sintered to gain high conductivity. The sintering of silver nanoparticles usually occurs between 160 °C and 210 °C [14]. The densification due to volume diffusion usually occurs between 300 °C and 350 °C [15]. Such low sintering temperatures are achieved because of the high surface area to mass ratio of nanoparticles [16]. This is beneficial for functionalizing low temperature materials, but can be unfavorable for higher temperature processes, like sintering of LTCC. The de-binding and sintering of the glass-ceramic material occurs beyond 350 °C, when the silver structures are already densified. Hence, the de-binding and shrinking due to sintering may lead to deformations in the densified silver structure. For screen printing, this problem is solved by LTCC shrinkage-adjusted pastes [1].

## 2. Methods

### 2.1. Tapes and laser process

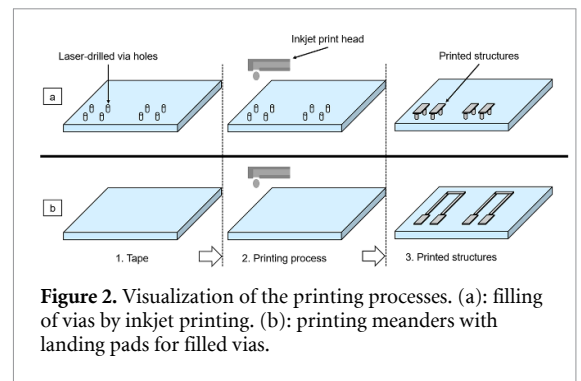
LTCC-tapes from DuPont, variant GT 951, were selected for the investigation with LTCC materials. The GT 951 tapes with thicknesses of 0.11 mm (PT), 0.165 mm (P2) and 0.265 mm (PX) were used for the further tests.

With a triple-pulsed 355 nm nanosecond UV laser, minimum via diameters of 40  $\mu\text{m}$  at the entrance hole and 30  $\mu\text{m}$  at the exit hole were achieved. The entrance of such a via is displayed in figure 5. Tapes with a thickness of 0.11 mm were used for the trials. Thicker tape variants had more glazing within the lasered openings and therefore cannot be filled adequately.

The 40  $\mu\text{m}$  vias were then arranged row by row in clusters of 2, 4 and 9 in a test layout with 6 rows and 8 columns to test the yield of the novel filling method. An example of a cluster of 9 vias is displayed in figure 6.

### 2.2. Printing processes

Inkjet-printing was performed on a DMP-2850 from Fujifilm Dimatix, USA in a clean room ISO 7. The silver nanoparticle ink I50T-13 from PV Nanocell, ISR was used as conductive ink. This ink has a rather low surface tension of 26  $\text{mN m}^{-1}$  to ensure that the wetting on LTCC is sufficiently high. I50T-13 was printed with 2.4 pL Samba and 10 pL DMC cartridges from Fujifilm Dimatix, USA. The 10 pL cartridges were utilized to print meanders. The 2.4 pL cartridges were susceptible to misfiring nozzles and were therefore only used to print pads and fill vias. Depending on the print head, drop spacings of 20  $\mu\text{m}$ , 10  $\mu\text{m}$  and 5  $\mu\text{m}$  were investigated. The suction of the tapes during printing was done by vacuum through porous paper. With this method, damage of the fragile tape



**Figure 2.** Visualization of the printing processes. (a): filling of vias by inkjet printing. (b): printing meanders with landing pads for filled vias.

due to the suction can be avoided and ink, which may flow through vias, will not reach the substrate table.

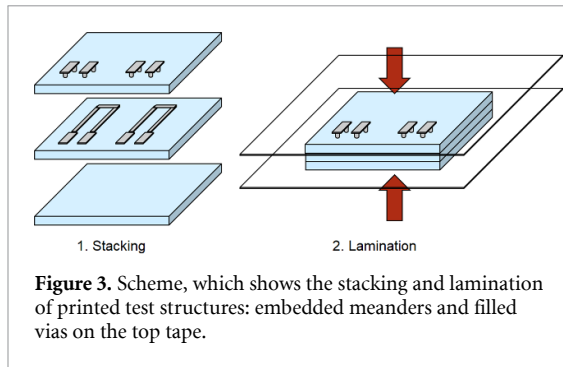
To investigate this new process for structuring LTCC, two layouts were chosen. One consists of meander structures with theoretical line widths of 100, 200, 300 and 400  $\mu\text{m}$ . At the start and end of the meander structures, landing pads for vias were included with a length of 4.5 mm and a width of 1.7 mm. The second printed layout was designed to fill vias. To fill vias, pads with the same dimensions as the meander layout were printed over vias.

In figure 2(a), the layouts and process steps for filling vias are shown. First, vias are laser drilled. After that, pads are inkjet-printed over the vias. The third picture shows the printed structures. For figure 2(b), the same steps for printing meanders on blank LTCC tapes are shown.

Two approaches for filling vias with inkjet technology are possible: filling vias before and after lamination of each layer. Since lamination is usually a time consuming process step, this publication does not deal with filling of vias after lamination. The goal is to be able to print on every tape separately, stack all tapes at the end of all printing processes and laminate only once after the tapes have been stacked. This process chain is more challenging, since ink can flow through the vias and wet the bottom of the tape, but it is much more appealing to production processes due to a reduction of process steps.

An alternative maskless process for filling vias is a squeegee process from the backside of a LTCC-tape. Since the backside is supported by carrier foil (so called Mylar foil), this is a maskless process, where vias are filled and then the supporting foil can be detached. For this process, a screen printing-compatible squeegee and a customized via-fill paste from DuPont were used. The silver paste was squeegeed over the pre-structured mylar foil into the punched openings of the LTCC tapes using a screen printer and the associated printing nest as well as an applied vacuum.

Printing on sintered LTCC was performed by inkjet and Aerosol Jet printing. Aerosol jet printing was performed on a M3D System from Optomec, USA. Silver nanoparticle ink I30EG-1 from PV



**Figure 3.** Scheme, which shows the stacking and lamination of printed test structures: embedded meanders and filled vias on the top tape.

Nanocell, ISR was used with a  $150\ \mu\text{m}$  nozzle diameter.

### 2.3. Stacking, lamination and sintering

The final LTCC for this paper consist of 3–4 tapes: one supporting base with a thickness of  $250\ \mu\text{m}$  and one tape with a thickness of  $160\ \mu\text{m}$  with inkjet-printed meander structures. The top tapes were either a tape with a thickness of  $110\ \mu\text{m}$ , with  $40\ \mu\text{m}$  vias, filled by inkjet printing and/or a  $250\ \mu\text{m}$  tape with  $250\ \mu\text{m}$  vias, filled by the described squeegee process.

After printing, the LTCC sheets were stacked with included stacking holes at the edges of each tape. After stacking, the LTCC were laminated in a hydraulic hot press RLKV 40/1 from Lauffer, Germany. The tapes were uniaxially laminated at  $100\ ^\circ\text{C}$  with 20–200 bar for 10 min. The schematic is presented in figure 3. Between the LTCC stack and the press, a separation foil was applied.

The laminates were then sintered at a maximum temperature of  $850\ ^\circ\text{C}$  with a 10 h sintering profile. The organic components of the tape and pastes were removed for 1 h at  $400\ ^\circ\text{C}$ .

### 2.4. Characterization

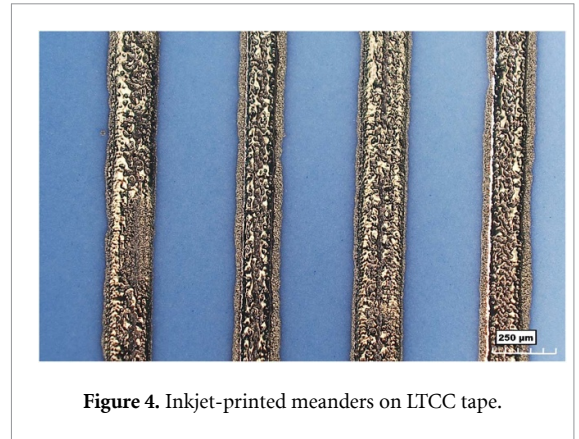
Printed structures were characterized optically by a laser autofocus system MLP-3 from Mitaka, Japan and light microscope RH-2000 from Hirox, Japan. Characterization by scanning electron microscope (SEM) was performed on JSM-6490LV from Jeol, Germany and FEI Helios Nanolab DualBeam 600 from Thermo Fisher, Waltham, MA, USA. Electrical characterizations were performed with the multimeter U1232A from Keysight, CA, USA. Embedded structures were analyzed by the x-ray microscope Cheetah from Comet Yxlon, Germany.

## 3. Results

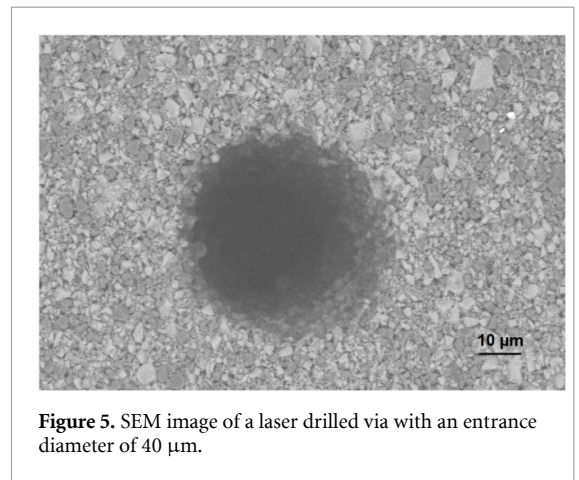
### 3.1. Printing on green LTCC tape

In a first step, meanders with theoretical line widths of  $100\ \mu\text{m}$ ,  $200\ \mu\text{m}$ ,  $300\ \mu\text{m}$  and  $400\ \mu\text{m}$  were printed. With a drop spacing of  $20\ \mu\text{m}$  and a 10 pl print head, continuous lines could be printed, see figure 4.

For the subsequent trials, meanders with theoretical line widths of  $100\ \mu\text{m}$  were selected. The mean



**Figure 4.** Inkjet-printed meanders on LTCC tape.



**Figure 5.** SEM image of a laser drilled via with an entrance diameter of  $40\ \mu\text{m}$ .

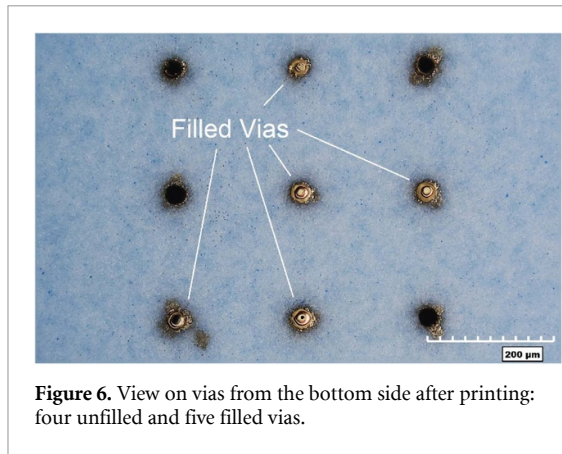
layer heights after drying at  $60\ ^\circ\text{C}$  were measured to be  $8\ \mu\text{m}$  for 3 printed layers with line widths of  $150\ \mu\text{m}$ . For inkjet printing on LTCC tapes, the number of printed layers was limited to 3. Besides the process time, another limiting factor is the soddening of the LTCC tape by the ink. Especially by printing without drying between layers and by printing on the  $110\ \mu\text{m}$  thin tape, the high amount of applied ink leads to soddening of the tape. This becomes problematic, because the tape can then adhere to the printing bed and cannot be removed.

To ensure that printed nanoparticle silver will not diffuse completely during high sintering temperatures of  $850\ ^\circ\text{C}$ , 3 layers were printed. Preliminary investigations with one printed layer showed that sintering of these thinner structures is possible, too. Such thin layers are however more susceptible to varying resistances, especially for smaller line widths.

### 3.2. Connection through vias

Initial filling tests were carried out with punched  $70\ \mu\text{m}$  and  $100\ \mu\text{m}$  openings, but these could not be filled safely. Ink passed through the vias and spread out at the via exit.

The lasered via holes were characterized by a SEM and a light microscope, see figure 5. Entrance diameters of the small vias varied between  $40\ \mu\text{m}$  and  $45\ \mu\text{m}$ . Exit diameters varied between  $25\ \mu\text{m}$  and  $27\ \mu\text{m}$ .



**Figure 6.** View on vias from the bottom side after printing: four unfilled and five filled vias.

**Table 1.** DoE for analysis of the influences of inkjet printing on the percentage shares of filled vias.

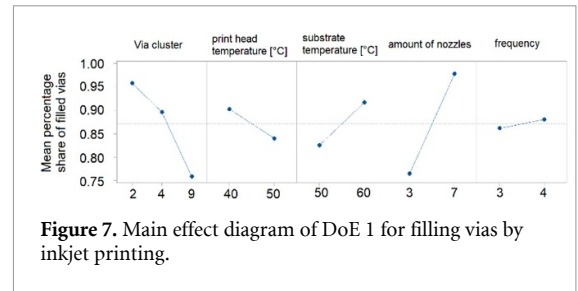
Factors	DoE 1—levels	DoE 2—levels
Via cluster	2, 4, 9	2, 4, 9
Print head temperature	40 °C, 50 °C	40 °C, 50 °C
Substrate temperature	50 °C, 60 °C	50 °C, 60 °C
Amount of nozzles	3, 7	1, 3, 5, 7
Printing frequency	3, 4	4

To quantitatively evaluate the amount of filled vias, the exit of each via on the backside of the structured LTCC tapes was analyzed after printing. One example is shown in figure 6 with 5 filled and 4 unfilled vias. These evaluation are further referred to as ‘percentage shares of filled vias’. For figure 6, the percentage share is 56%.

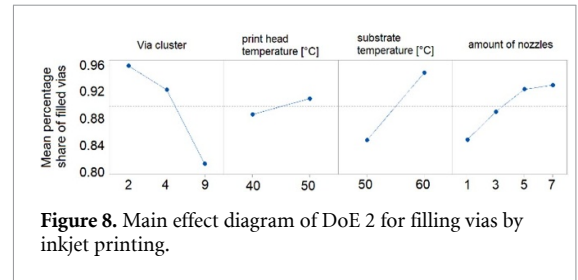
In first trials for filling 40  $\mu\text{m}$  vias, varying percentage shares of filled vias, depending on the printing parameters were observed. To investigate the reasons for the varying percentage shares of filled vias, two full factorial design of experiments (DoE) were conducted to gain insights into the most important processing factors. Even parameters, which are typically varied between print jobs, like the number of active nozzles, were investigated thoroughly. The parameters of both DoE are listed in table 1. For clusters of 2, 4 and 9 vias, the sample sizes for each level were 16, 32 and 72. The vias were filled with 2.4 pl cartridges and a drop spacing of 10  $\mu\text{m}$ .

The results of the first full factorial DoE are presented in figure 7. It turns out that especially a higher number of nozzles and a smaller number of vias significantly benefit the percentage share of filled vias. A high substrate temperature of 60 °C seems to have a positive influence on the amount of filled vias as well.

To gain more insights, the second DoE was performed with more levels of used nozzles. The experiments were conducted in a randomized order to ensure the validity and reliability of the results. This second DoE confirmed that the cluster of 2 vias, a higher substrate temperature and a higher number of



**Figure 7.** Main effect diagram of DoE 1 for filling vias by inkjet printing.



**Figure 8.** Main effect diagram of DoE 2 for filling vias by inkjet printing.

nozzles benefit the percentage share of filled vias, see figure 8.

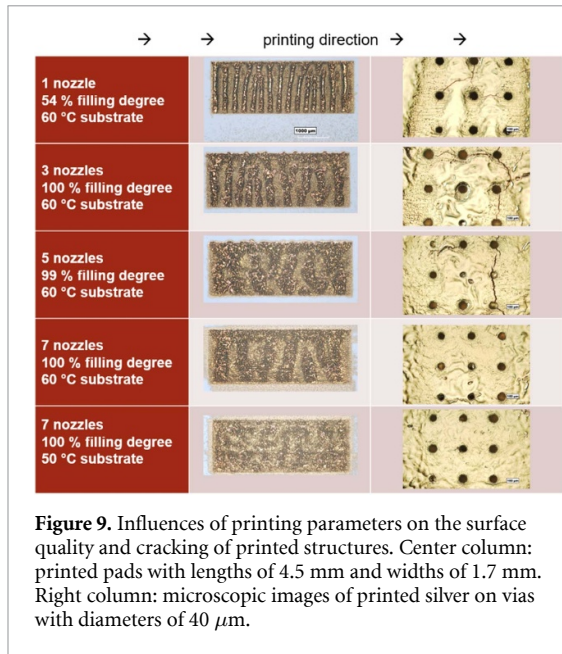
Besides the percentage share of filled vias, the surface quality of the printed structures and the susceptibility towards cracking during drying were analyzed. Optical analysis of printed structures with substrate temperatures of 60 °C are summarized in figure 9. The pictures in the central column show printed pads with a length of 4.5 mm and a width of 1.7 mm. Extreme drying patterns can especially be seen on the structures, which were printed with only one nozzle. This printing strategy results in periodically aligned accumulations of nanoparticles, perpendicular to the printing direction. It seems that a high number of nozzles benefits the homogeneity of the printed structures. Pads, printed with seven nozzles, showed a uniform distribution of ink. On the right column, close-up microscopic images of ink, printed on vias, are displayed. These images show that cracks rather appear on structures, printed with one nozzle, than structures printed with seven nozzles. Those results were confirmed for trials at 50 °C.

### 3.3. Lamination

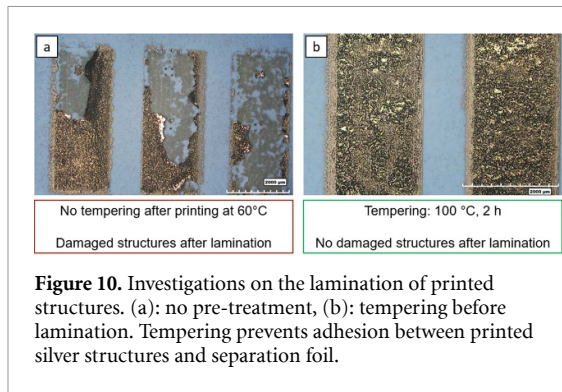
The lamination of printed LTCC tapes was first investigated without further treatment of the printed structures, besides drying at 60 °C on the printing bed. Without any further treatment, the printed structures on the top layer can partially adhere to the separation foil, see figure 10(a). Since the lamination process is performed at 100 °C, a drying step at 100 °C was performed prior to the lamination in another trial. With this process step, adhesion between the printed structures and the separation foil was completely prevented, see figure 10(b).

### 3.4. Co-firing

Co-firing was successfully performed at a maximum temperature of 850 °C. Embedded, sintered meanders



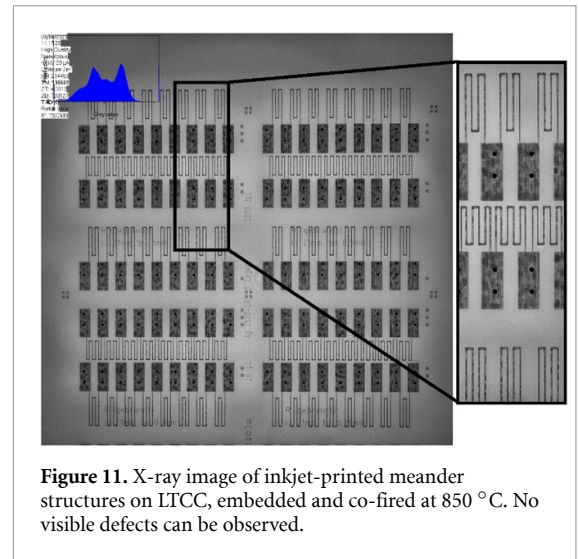
**Figure 9.** Influences of printing parameters on the surface quality and cracking of printed structures. Center column: printed pads with lengths of 4.5 mm and widths of 1.7 mm. Right column: microscopic images of printed silver on vias with diameters of 40  $\mu\text{m}$ .



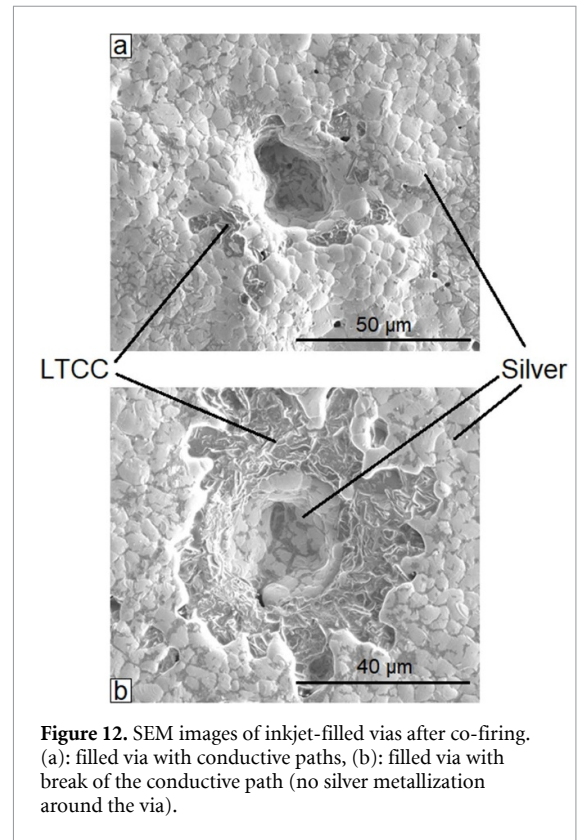
**Figure 10.** Investigations on the lamination of printed structures. (a): no pre-treatment, (b): tempering before lamination. Tempering prevents adhesion between printed silver structures and separation foil.

were analyzed by measuring the electrical resistances. For those trials, meanders were laminated between a support tape and a top tape, with vias, filled by a squeegee-based process. Prior to lamination, the dimensions of printed meanders were measured. The line widths, heights and lengths were 150  $\mu\text{m}$ , 8  $\mu\text{m}$  and 20 mm. The connection from top pad to top pad on the LTCC component, which includes vias and one embedded meander, showed a mean electrical resistance of 0.67  $\Omega$  with a standard deviation of 0.03  $\Omega$  after co-firing, for 96 embedded samples, with all samples being conductive. An x-ray image in figure 11 proves the quality of inkjet-printed meanders after co-firing at 850  $^{\circ}\text{C}$ , since no visible defects can be observed. The same trials were performed on LTCC-stacks with an additional tape with inkjet-filled vias, between the squeegee-filled top tape and the inkjet-printed meanders. These 96 structures were only conductive in 28%–37% of the samples with a mean electrical resistance of 1.44  $\Omega$  with a standard deviation of 0.98  $\Omega$ .

To investigate the low amount of conductive, inkjet-filled vias, another trial was performed: Inkjet filled vias on top tapes were co-fired at 850  $^{\circ}\text{C}$ . After



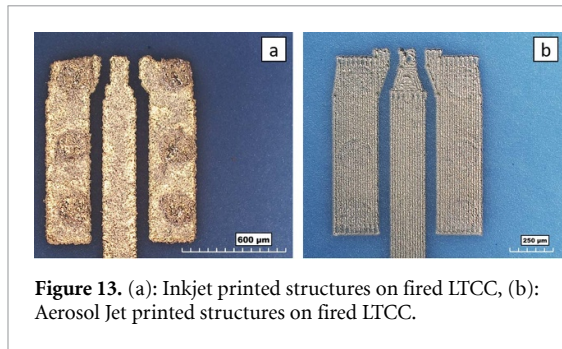
**Figure 11.** X-ray image of inkjet-printed meander structures on LTCC, embedded and co-fired at 850  $^{\circ}\text{C}$ . No visible defects can be observed.



**Figure 12.** SEM images of inkjet-filled vias after co-firing. (a): filled via with conductive paths, (b): filled via with break of the conductive path (no silver metallization around the via).

co-firing, they were analyzed by SEM, displayed in figure 12. Figure 12(a) shows a via, filled with silver and only a few gaps in the silver structure. The via is covered with silver and the conductivity to the rest of the silver is given. Figure 12(b) however shows silver inside of a via, but no connection to the rest of the silver on the top tape of the LTCC. The low amount of silver on the edges seems to be the reason for such a non-conducting via.

Sintered top layers were analyzed by microscopic images. The printed pads showed blistering of the silver structures. This could be avoided by printing on the top layer of the already co-fired LTCC



**Figure 13.** (a): Inkjet printed structures on fired LTCC, (b): Aerosol Jet printed structures on fired LTCC.

and subsequently sintering the printed structures at 200 °C–600 °C, which is described in section 3.5.

### 3.5. Printing on co-fired LTCC

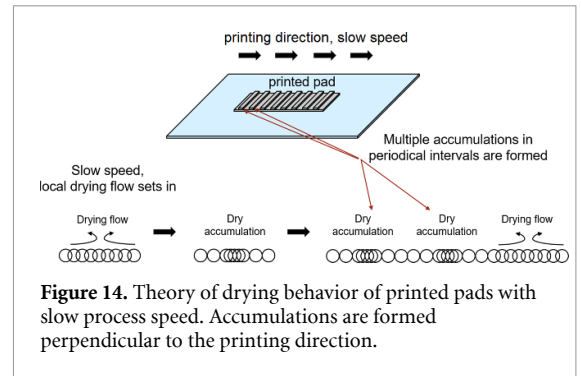
Printing on fired LTCC was investigated by inkjet and Aerosol Jet. Figure 13 shows examples for both technologies. Both printing layouts were optimized to accurately print the designed CAD file. For Aerosol Jet printing, the achieved line widths and pitches were reproduced within a range of  $\pm 10 \mu\text{m}$ . For inkjet printing, such an accuracy was not achieved across the whole LTCC board. Variations in wetting resulted in various shorts, especially for fine pitches.

Sintering was performed at 200 °C, 300 °C, 400 °C and 600 °C. The goal of the varying sintering temperatures was to investigate the influence on the adhesion between the printed structures and the LTCC. The adhesion was observed during cutting of the LTCC. During cutting with a water-cooled wafer saw, Aerosol Jet printed structures partly detached from the LTCC board, for sintering temperatures of 200 °C, 300 °C and 400 °C. Samples, sintered at 600 °C, withstood the cutting process. They also withstood a tape test with a tape with adhesive strength of  $2.5 \text{ N cm}^{-1}$ . A meander with a length of 600 mm, mean width of  $120 \mu\text{m}$  and mean height of  $3.3 \mu\text{m}$  shows a specific electrical resistance of  $2.8 \mu\Omega\text{cm}$ , which is around 57% of the bulk resistivity of silver.

## 4. Discussion

Printing on LTCC with the silver ink I50T-13 with 10 pL DMC and 2.4 pL Samba cartridges worked well. The surface tension of the ink of  $26 \text{ mN m}^{-1}$  is low enough to achieve good wetting with a drop spacing of  $20 \mu\text{m}$  for the 10 pL cartridges and a drop spacing of  $10 \mu\text{m}$  for the 2.4 pL cartridges. With the used settings, line widths of  $150 \mu\text{m}$  were printed. After sintering, 100% of the 96 printed meanders were conductive, which shows that inkjet printing of conductive tracks can be performed well on LTCC and co-firing at 850 °C can be performed with embedded printed nanoparticles.

Investigations of inkjet-printed pads reveal that printing with only one nozzle can lead to inhomogeneous drying and even cracking. A theory for this phenomenon is that slow process speed, resulting of one

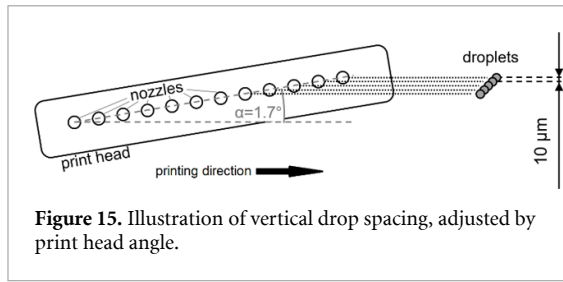


**Figure 14.** Theory of drying behavior of printed pads with slow process speed. Accumulations are formed perpendicular to the printing direction.

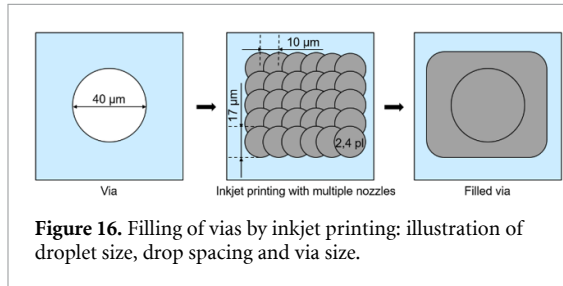
active nozzle and a frequency of 4 kHz, can result in local drying effects during the printing of a pad. This is even more accelerated by a rather high printing bed temperature of 60 °C, which accelerates the evaporation of the solvent. The theory behind the observed accumulations is displayed in figure 14. The accumulations may be formed perpendicular to the printing direction, because the local drying effects are faster, than the printing of a full swath. This is comparable to the stacked coin effect, which could occur if the jetting would be even slower or the substrate temperature would be even higher [17]. If the number of nozzles increases, the amount of ink, deposited during one pass increases as well. With more ink, the time needed for drying increases. Furthermore, the accumulations should have a wider footprint. This behavior was observed in figure 9. If the pads are printed with 7 nozzles and a lower substrate temperature of 50 °C, no accumulations can be found and the desired homogeneity of printed pads can be achieved.

Two full factorial DoE showed that filling of vias with multiple nozzles is beneficial, in order to fill vias down to their exit. Printing with only one nozzle can lead to the observed behavior of only partly filled vias. One reason for this can be the spreading, contraction and drying of a droplet, after the impact [18, 19]. If the contact line of the droplet recedes after the initial spreading, the droplets may dewet from the bottom of the via. This effect may occur if the vias are not filled completely in one pass. In that case, the droplets have enough time to recede and dry, before the next line is printed. The nozzles of the used Samba cartridge are aligned in one line. The drop spacing, perpendicular to the printing direction, is adjusted by the angle of the print head. For 5 active nozzles, 5 lines of droplets are printed with a drop spacing of  $10 \mu\text{m}$ , by setting the print head angle to  $1.7^\circ$ , see figure 15. With multiple lines printed over a via within one pass, the chance of fully covered vias increases, which is illustrated in figure 16. This may inhibit the dewetting in the via and lead to filled via-exits.

Sintering of inkjet-filled vias was investigated by SEM. It can be seen that silver on the top side of the vias cannot always be found after sintering. Gaps in the silver structure can especially occur on the edges of the entrance diameter. To evaluate the sintering of



**Figure 15.** Illustration of vertical drop spacing, adjusted by print head angle.



**Figure 16.** Filling of vias by inkjet printing: illustration of droplet size, drop spacing and via size.

inkjet-filled vias quantitatively, different LTCC setups were tested. The first setup contained one tape with inkjet-printed meanders, one layer of inkjet-filled vias and one layer of vias, filled by a squeegee process. Those were compared with LTCC setups with one layer of inkjet-printed meanders and one layer of vias, filled by a squeegee process. Electrical characterizations show that only 28%–37% of the samples with inkjet-filled vias were conductive after sintering. On the other hand, 100% of the samples without inkjet-filled vias were conductive after sintering. This shows two things: On the one hand, the squeegee-based process seems to be well suitable for a maskless production of LTCC. On the other hand, filling vias in LTCC tapes with the current inkjet printing process chain cannot be recommended. For future research on inkjet-filled vias, improvement of the wetting on the edges of the vias can be promising to achieve a higher percentage share of conductive vias. This can result in a more homogeneous thickness of the printed silver, which can result more continuous silver layers after sintering. The wetting may vary on the edges due to the laser drilling process, which induces heat and can lead to rough edges. A picosecond pulsed laser can be promising to drill smoother edges for an improved wetting of the ink.

During co-firing at 850 °C, deformations in printed silver structures on the top layer were observed. Since the sintering of printed nanoparticles typically occurs below 350 °C, the sintering of nanoparticles occurs early in the co-firing process. The deformations seem to be especially problematic for printed structures on the top layer of LTCC, because there are no constraints to prevent the deformation of silver structures. In the LTCC embedded state, both the symmetrical structure of LTCC layers to the printed structure and the reduced oxygen absorption of the silver tracks prevent the deformation of the printed structures [20]. Hence, printing with a process chain,

in which printing and sintering of the top layer is only performed, after the co-firing process is already finished, can be recommended.

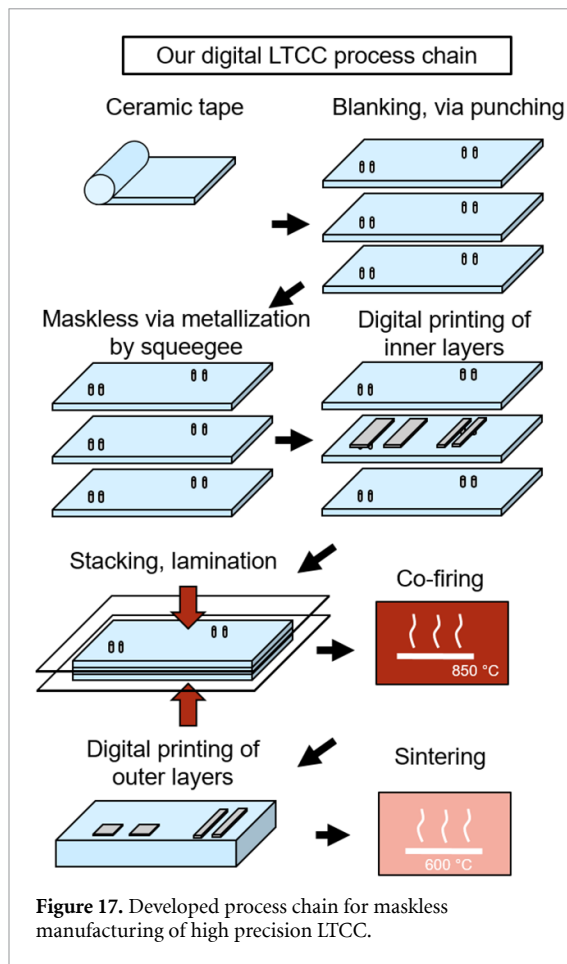
Printing on the top layer was investigated regarding accuracy and adhesion. For structures, which require high accuracy, Aerosol Jet printing was utilized. With this rather dry digital printing process, no fluctuations of the wetting were observed. The CAM data were adjusted to fit the CAD data. With the optimized CAM data, variations below  $\pm 10 \mu\text{m}$  of the printed layout compared to the CAD data were achieved. Hence, this process can be very promising for high frequency applications [5]. For structures, which do not require accuracies below  $\pm 10 \mu\text{m}$ , inkjet printing was utilized. Variations in the accuracy across the fired LTCC board were observed for inkjet-printed structures. Such variations can be caused by misfiring nozzles, varying surface tension across the ceramic test board or high surface roughness [21]. These variations can result in shorts, especially with small pitches below 100  $\mu\text{m}$ . Low roughness, optimized ink, cleaning and plasma routines may result in optimized wetting across the whole board and can be investigated in the future [21, 22].

The adhesion of the printed structures was observed after sintering at 200 °C, 300 °C, 400 °C and 600 °C. Only the adhesion of the printed structures, sintered at 600 °C, was high enough to withstand the vibrations and impact of water-cooling during wafer sawing of the LTCC components. Silver is prone to diffusion into LTCC [23, 24]. The diffusion rate at 600 °C may be high enough, that printed nanoparticles can partly diffuse into the LTCC, which improves the adhesion. Hence, for mechanically loaded structures, sintering printed nanoparticles on LTCC at temperatures of 600 °C are recommended.

With the developed processes, a maskless process chain for manufacturing LTCC can be presented. Vias are filled from the backside of the tape by squeegeeing silver paste over punched LTCC and mylar foil. This is the only process in the process chain, which requires an additional printer and can currently not be performed by inkjet printing. However, maskless processes can be performed by this approach and companies which own such printers can eliminate the utilization of screens. This will enable faster R&D cycles since screens do not need to be fabricated for each LTCC layer and also costs can be saved for small scale production.

Further conductive structures are applied by inkjet printing. Then the tapes are laminated and co-fired. Afterwards, the top layer is structured by digital printing processes like inkjet or Aerosol Jet and subsequently sintered at 600 °C. Both approaches show good printing results, however Aerosol Jet printing shows the better reproducibility for narrow tolerances throughout a LTCC board. The process chain is illustrated in figure 17.





## 5. Conclusion

Inkjet printing of silver nanoparticle ink on tapes for LTCC was demonstrated in this paper. The tapes with printed structures were stacked, laminated and co-fired at 850 °C. After co-firing, 100% of the embedded inkjet-printed structures were conductive and no visible defects were observed. This shows the good suitability of inkjet printing for functionalizing of LTCC tapes.

Filling of vias by inkjet-printing was investigated. The most important parameters for filling of vias were found to be the number of active nozzles, substrate temperature and the type of via clusters on the substrate. After co-firing at 850 °C, 28%–37% of embedded via clusters were conductive. An alternative, maskless, squeegee-process was utilized to produce 100% conductive via clusters.

After LTCC tapes were structured with maskless technologies, laminated and co-fired, the top layers were structured by inkjet and Aerosol Jet printing. With those technologies, highly accurate conductive structures were produced.

The presented process chain is very promising for small scale production of LTCC or research and development processes. The small and accurate feature size of the printed structures on fired LTCC can also be promising for application areas like high

frequency technology. Future research can focus on further optimization of filling of vias by inkjet printing, further miniaturization as well as investigating the properties regarding specific applications.

## Data availability statement

The data that support the findings of this study are available upon request from the authors.

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