Full paper

Toward 3D-Printed Electronics: Inkjet-Printed Vertical Metal Wire Interconnects and Screen-Printed Batteries

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Inkjet and screen printing technologies are well known in the graphic arts industry for the reproduction of texts, images, and graphics. During the last decades, these printing technologies have been attracting increasing interest for the deposition of functional materials, e.g., in the field of printed electronics and for biological applications. However, their usage is mainly limited to 2D applications, i.e., rather flat deposits ranging from nanometers to several tens of micrometers in thickness. For 3D applications, sophisticated additive manufacturing technologies are developed to manufacture structures with high shape complexities. Herein, the potential of standard inkjet and screen printing technology as tools for the development of functional 3D objects is demonstrated. 3D functional structures printed by inkjet and screen printing technology combining conductive and nonconductive materials to a multi-material structure are shown. A metal nanoparticle ink formulation is applied to inkjet-print conductive metal pillars with a high aspect ratio (in the range of 50) used as vertical interconnects. The interconnects are encapsulated with an inkjet-printed polymer ink formulation and finally used as conductive tracks to light up a solid-state light-emitting diode (LED). Screen printing is applied to print primary batteries used as the power source for the LED.

1. Introduction

The term 3D printing encompasses a large range of technologies and processes that have usually a common basic principle: additive layer-based manufacturing. Today, there are many 3D printers commercially available allowing the processing of materials such as polymers, ceramics, metals, tissues, wax, etc. at different scales.[13–7] However, although a lot of development has been done in 3D printing, it is still difficult to combine different materials in a single 3D object using a single 3D manufacturing process. One of the key benefits of 3D printing is the possibility to create highly complex shapes and structures, e.g., with internal cavities, which is quite difficult to manufacture with traditional methods. Screen printing or inkjet printing is usually not considered as a 3D printing technique—although both of the technologies are additive layer-based technologies.[8] Inkjet printing is a versatile technology finding increasing application for the field of printed electronics, e.g., printing of electronic devices such as inductors,[9] capacitors,[10] diodes,[11] sensors,[12] memories,[13] and transistors.[14] These devices are built by thin inkjet-printed layers of different conductive[15,16] and nonconductive materials, stacked on top of each other. Thus, these thin-film electronic devices could be considered as tiny 3D objects in the micrometer to millimeter range although the thickness of the devices is significantly lower than their lateral dimensions. In this research contribution, we demonstrate the potential of inkjet printing technology for the manufacturing of 3D objects with layer thicknesses significantly higher than their lateral dimensions. In detail, conductive 3D objects are printed having a layer thickness close to the centimeter range but a lateral size of only some hundreds of micrometer. These objects can be used as vertical interconnects for various electronic applications. Different methodologies are known for the preparation of vertical interconnects between isolated stacked layers on, e.g., a printed circuit board (PCB) board via hole fabrication in dielectric materials and the filling of these via holes with a conductor[17] or the preparation of vertical, conductive pillars with their subsequent embedding/encapsulation in a polymer resin. The first method needs either additional subtractive steps for making vias, or highly sophisticated manufacturing technologies for the generation of the layer directly including vias avoiding the subsequent subtractive via preparation. The uniform filling of
vias with a depth in millimeter range is very challenging employing traditional technologies or printing technologies, e.g., due to material shrinking effects as shown by Khorramdel and Mäntysalo. Therefore, many researchers have focused on the manufacturing of vertical interconnects without a predefined via.

In this research work, 3D multilayer interconnectors by inkjet printing with potential application in electronic devices, e.g., PCBs, were studied. In comparison to chip-to-socket micro connectors, electronic components of PCBs can be placed within horizontal or vertical distances in several hundred micrometer, millimeter, or even centimeter range. For these applications, we demonstrate the potential of inkjet printing to manufacture metal vertical interconnects of varying height. To indicate the high flexibility of inkjet printing, the metal vertical interconnects were embedded in an inkjet-printed, UV-cured polymer block. The top surface of the polymer block is used as a “second height level” to demonstrate the proper function of the 3D interconnects used as conductive tracks to power a light-emitting diode (LED).

The research about the application of inkjet printing technologies for the preparation of 3D interconnects/pillars is still in progress and several studies are found in the scientific literature. Different materials were used in these studies such as metal nanoparticle inks or molten metal, inks with the conductive polymer PEDOT:PSS, and also nonconductive materials such as ceramic particles formulated to inks. Droplets containing the aforementioned materials were ejected by inkjet printheads and stacked on each other, thus forming a layer with layer height increasing orthogonally with that of the substrate. The final 3D object is usually called as vertical pillars if the diameter of the objects is in the range of tens of micrometers, thus corresponding roughly to the diameter of a single droplet. Bhatti et al. demonstrated printed ceramic pillar arrays on a siliconized paper with a modified drop-on-demand piezo inkjet printer from IBM. The pillars were printed with 4000 layers and had an average height of 1.3 mm and width of 120 μm. Also, Zhao et al. was printing ceramic pillar arrays using inkjet printing. The pillars were printed with 4900 layers and had an average height of up to 2 mm and widths of 65 μm. de Gans et al. studied inkjet printing of polymers taking in account 3D-printed polymer objects. Ko et al. fabricated different 3D microstructures with complex geometry using a gold nanoparticle inkjet ink. Kullmann et al. investigated the influence of the temperature of the substrate on the morphological and electrical properties of the printed gold pillars and manufactured pillars with a height up to 7 mm. Sadie and Subramanian analyzed the influence of different printing and sintering parameters on electrical, mechanical, structural, and morphological properties of fabricated microstructures. An et al. demonstrated the preparation of pillars and different structures with complex geometry using silver, copper, and cobalt as well as anthracene and 6,13-bis(trisopropylsilyl)ethynyl)-pentacene (TIPS-pentacene) inks by electrohydrodynamic inkjet printing to be applied for highly integrated devices in next-generation electronics.

Next to inkjet printing technology, many other approaches such as chemical and physical vapor deposition as well as flow dispenser systems were studied as well to manufacture pillars. Detailed information about the state-of-the-art of inkjet printing of functional 3D pillars can be found as a comprehensive summary in the Supporting Information.

The manufacturing of thin-film batteries employing printing technologies has been studied for about already about two decades. The development history of screen-printed primary batteries is shown in a snapshot by Willert and Baumann. The state-of-the-art of printed batteries on both research on industrial level is reviewed in 2015 by Sousa et al. and in 2018 in a very detailed manner by multiple authors in the study by Lancers-Méndez and Costa. While the rechargeable types of batteries such as NiMH or lithium-based systems require aggressive alkaline electrolytes, primary batteries can work with an ecological mild aqueous ZnCl₂ electrolyte and therefore can be called “green technology.”

Many different applications for printed batteries have been addressed during the last years, e.g., printed batteries were applied for the energy supply of a cholesterol sensor. Currently, there are also temperature tracker patches from Blue Spark Technologies (USA) commercially available that are built up with printed primary zinc carbon batteries. Also, many other scientists and companies aim to commercialize the well-known zinc-carbon system, e.g., the Changzhou Institute of Printed Electronics Industry in China. In 2018, the application of printed batteries on technical textiles has been demonstrated contributing toward the vision of smart textiles. Another approach for printing batteries is maintained from Evonik and called “Taettooz.” This development is based on activities of The Schubert Group in Jena, Germany, over the last decade. In these batteries, a polymer-based rechargeable battery is built. The electrolyte is UV curable and, therefore, a solid layer system can be deposited by screen printing. The only restriction compared with a primary zinc-carbon system is that the area capacity is about one order of magnitude lower.

2. Experimental Section

Figure 1A shows schematically the architecture of the inkjet-printed 3D demonstrator. It consists of two bottom contact electrodes inkjet-printed on a glass substrate, and two vertical pillars inkjet-printed on top of the bottom electrodes. The vertical pillars were embedded inside an inkjet-printed polymer block (polymer body). Two dispersed top electrodes connect the ends of the vertical pillars with a light-emitting diode (LED) on top of the polymer block to prove the functionality of the printed demonstrator. A screen-printed battery was connected to the bottom electrodes on the glass substrate to supply the LED with power. A schematic architecture of the screen-printed battery with all the different materials is shown in Figure 1B.

Standard float glass microscope slides (76 × 26 mm area size, 1 mm thickness) were applied as substrates. Before printing, dirt and dust particles were wiped off from the glass slides using ethanol impregnated clean room tissues. After that, the glass slide was rinsed with ethanol. Finally, the substrate was dried in a nitrogen flow for few seconds and conditioned in the printer for about 5 min.

Two commercially available ink formulations for inkjet printing were applied. The bottom electrodes and vertical pillars were printed using the silver nanoparticle ink NPS–JL (Harima...
Chemicals Group Inc., Japan). The nanoparticles have a particle size of about 7 nm, a metal content of 52–58 wt%, an ink viscosity of 11 cP, and the specific electrical resistance of a printed layer after sintering is about 0.06 Ω mm²/cm² (drying and sintering for 60 min at 120 °C is recommended by the ink manufacturer, all data according to the ink manufacturer). The polymer body was inkjet-printed using the UV-curable polymeric ink formulation V Photon Clear NonWet (Tritron GmbH, Germany). The top electrodes connecting the top ends of the vertical pillars with the LED on the polymer body were made with a standard conductive silver lacquer by manual dispensing.

Inkjet printing was carried out with the well-known drop-on-demand inkjet printing system Dimatix Materials Printer 2831 (DMP2831, Fujifilm Dimatix Inc., USA). The printing system was equipped with a laboratory cartridge printhead DMC-11610 (16 nozzles, ≈21.5 μm nozzle diameter, 10 pL nominal drop volume). All 16 nozzles were selected for printing at a maximum jetting frequency of 1 kHz. Further information about the waveform and drop ejection of the printhead with the silver ink is shown in Supporting Information (Figure S1, Supporting Information). Additionally, the printing system was equipped with the inline UV cure system PEL Dual UV (Printed Electronics Ltd. [PEL], United Kingdom) that allows UV pinning and curing during the printing process. All 16 nozzles were selected for printing at a maximum jetting frequency of 1 kHz. Further information about the waveform and drop ejection of the printhead with the silver ink is shown in Supporting Information (Figure S1, Supporting Information). Additionally, the printing system was equipped with the inline UV cure system PEL Dual UV (Printed Electronics Ltd. [PEL], United Kingdom) that allows UV pinning and curing during the printing process. The cure system is based on an LED with the peak wavelength of 365 nm, a focal length of 3 mm, a maximum output power of 500 mW, and an energy density of ≈8500 mJ cm⁻² (all parameters according to the manufacturer). Geometrical sizes and the microstructure of the printed vertical pillars were analyzed with the scanning electron microscope (SEM) Hitachi TM-1000. Electrical measurements were carried out using the digital multimeter Tektronix DMM4040 connected to the measurement probe station PM5 from Suess MicroTec.

The manufacturing and post-treatment settings as well as detailed information regarding digital printing patterns and inkjet manufacturing sequence are given in Supporting Information (Table S2, Figure S2, Supporting Information). First, the bottom electrodes were printed with silver nanoparticle ink on the prepared substrate and sintered on a hot plate. The nozzle-to-substrate distance was set to 1 mm. Second, 100 identical layers were inkjet printed on top of each other with the nanoparticle silver ink to develop the vertical pillars on top of the bottom electrodes. The nozzle-to-substrate distance for the pillar printing process was set to 5 mm. After the printing, the pillars were sintered for 10 h in a hot air oven at 100 °C. Third, the outer contour of the polymer body was printed using the Tritron ink with 100 identical layers stacked on top of each other with UV curing during the printing process. After the dielectric printing, the previously printed vertical silver pillars are placed freely inside the printed polymer contour. Fourth, the printed polymer contour was exploited as a vessel and filled using inkjet printing with the Tritron ink and UV-cured with the PEL LED. The filling was done in such a way that the conductive silver pillars protrude with about 0.5 mm from the polymer body. Finally, two top electrodes using the silver lacquer were deposited manually on top of the polymer body and connected to the protruding...
silver pillars and an LED. The LED has a green light and was ordered from Kingbright made with AlGaInP on GaAs substrate \((3.2 \times 1.6 \text{ mm}, \text{ thickness } 1.8 \text{ mm, part number KPTD-3216CGCK})\). Finally, the screen-printed battery was connected to the free ends of bottom electrodes on the glass slide to close the electrical circuit.

The screen-printed battery was built on a polyethylene terephthalate (PET) film (Melinex 401 CV 100 \(\mu\)m). The current collector was based on a carbon ink. Homemade zinc and manganese dioxide inks were used for the two electrodes. A gelled aqueous \(\text{ZnCl}_2\) electrolyte has been employed together with a fiber-based separator. Double-sided glue tape was applied in the laminating process to encapsulate the entire battery system except two electrodes with the PET substrate. The battery layers were printed with an EKRA E1-XL screen printing machine and dried by hot air in a 2 m drying zone using a 3D-Micromac AG microDRY machine. The temperature was kept below 110 °C to avoid any damage at the PET film.

3. Results and Discussion

Figure 2 shows the inkjet-printed conductive pillars as photograph as well as SEM images. The SEM images show three different pillar heights; about 430, 1100, and 2200 \(\mu\)m. The variation in height was obtained by a simple variation of the number of layers (24 layers, 48 layers, and 96 layers, respectively). Printing of one layer only forms a flat deposit as demonstrated in Figure 2B. The three printed pillars with different layer numbers shown in Figure 2C,D,E have a similar shape as pillars of other studies and consist mainly of three structural elements: 1) large pillar basement formed on top of the substrate (highest lateral extension of the pillar); 2) main pillar body with typical ring-like sections as result of the printed inkjet droplets; and 3) smooth hemispherical pillar vertex.\(^{[21]}\) Figure 2F shows the cross-section of a pillar that has been detached from the substrate. Directly after printing, the pillars are usually straight without any bending and with a smooth surface as shown in Figure 2G. Drying and sintering were carried out at 100 °C for 10 h in oven and is causing a shrinkage of the pillars. The shrinkage results in a physical deformation of the pillar surface that is quite obvious when comparing Figure 2G (as printed) with Figure 2C,D,F,H (sintered). A drying and sintering for a shorter period but at >100 °C was causing even more intense physical deformations of the pillars. In some cases, the pillars were broken. The pillar height increases proportionally with the printed layer number. Mainly due to the coalescence of the densely deposited ink droplets and as a result of the interplay between surface tension of the ink and surface energy of the substrate, the pillars form a cylindrical shape although the digital printing pattern applied was square-shaped. In comparison to other studies where only one ink droplet per layer was deposited, in our case many droplets were used per layer. Therefore, the ink remains liquid for a longer time and can spread over the substrate or over the previously printed layers. During the spreading, the shape adopts an energy-effective form that is ideally a sphere. This fact can explain the cylindrical body form as well as the hemispherical vertex form of the pillars and also the increased pillar diameter (\(\approx 200 \mu\)m diameter for printed pillars although 120 \(\mu\)m were defined as width in the digital pattern). The printing process shows a stable vertical growth of the manufactured pillars with increasing layer count. The maximum achieved pillar height was \(\approx 10 \text{ mm} \) (Figure 2A) with 200 printed layers and an aspect ratio of 50:1. Also, higher pillars might be possible.

Since the inkjet printer allows only the deposition of one material at the same time (only one printhead with one channel can be installed), the deposition of the polymer block has to be done after the printing and sintering of the silver pillars. However, printing the polymer block around the silver pillars is challenging due to the high distance required between the printhead and the substrate (basically height of the vertical pillar plus 500 \(\mu\)m safety distance). The UV lamp was installed at a similar height as the printhead causing a high decrease of energy density at the layers printed close to the substrate. The polymer block was printed with repeating digital rectangular patterns of the same size and position. However, as shown in Figure 3A,B we did not obtain a cuboid shape but a kind of truncated pyramid shape. The
reason is most probably the insufficient energy density of the UV radiation allowing a higher ink spreading of the layers located closer to the substrate in contrast to the layers located closer to the UV lamp. However, despite the deformed shape, one can see quite well the pillars embedded in the yellowish polymer material protruding on the top of the polymer block. To solve the problem of the deformed shape, only the contour of the rectangle was inkjet-printed and UV-cured but not the inner part. With that method, a lower ink volume per layer count was obtained. With increasing number of layers, the contour of the rectangle increases in height and a vessel is formed, which was subsequently filled up with ink by inkjet printing. Finally, the inner printed part was UV-cured. With this approach, the obtained polymer block has a cuboid shape with a flat surface and the previously manufactured silver pillars were embedded reliably (Figure 3C).

Before performing the printing of the polymer body contour, different tests were carried out to study the influence of printing parameters on the contour formation, e.g., a simple line pattern consisting of lines with a length of 20 mm and varying line widths from 0.1 to 1 mm in steps of 0.1 mm and additionally a line pattern with a length of 20 mm and a width of 1.5 mm. Two hundred layers of the line pattern were printed on top of each other at 1693 dpi (UV curing was done with the PEL UV system while printing) and with a nozzle-to-substrate distance of 5 mm. Figure 4A shows the obtained 3D sample based on the inkjet-printed line pattern. Small 3D objects looking like small walls were formed by inkjet printing using the UV curable ink formulation. Obviously, very different morphologies of the printed layers were obtained although the printing resolution, the number of layers, and the line length were identical for all line widths. These results indicate that the line width influences dramatically the shape and especially the height of the printed wall deposits. Figure 4B shows the geometrical data of the printed lines as a function of the digital line width. Obviously, lines printed with a digital line width < 0.6 mm have a quite constant measured line width that is also much higher than the digitally set line width. The measured line widths of the lines printed with a digital line width of > 0.6 mm are more close to the digitally set line width. In contrast, the line height dramatically increases for digital line widths < 0.7 mm and has a smaller slope of increase for digital line widths > 0.7 mm. This behavior is a function of ink spreading and wetting. The deposited ink volume was approximated by multiplication of the ink droplet volume with the number of deposited ink droplets per line. The optically determined ink droplet volume was about 10.8 ± 0.7 pL. The approximated deposited ink volume was plotted to compare it with the measured printed material volume. The measured printed material volume was determined by a simple geometrical

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**Figure 3.** A) Side view and B) angular view of the inkjet-printed polymer body forming a kind of truncated pyramid shape printed with the V Photon Clear NonWet ink (printing resolution, 2540 dpi; nozzle-to-substrate distance, 10 mm; 100 layers) around the silver pillar printed before. C) Polymer body obtained by printing first the contour and subsequently filling the contour with the UV ink. All samples were printed with the V Photon Clear NonWet ink.

**Figure 4.** A) Inkjet-printed polymer walls (V Photon Clear NonWet ink; printing resolution, 1693 dpi; nozzle-to-substrate distance, 5 mm; 200 layers; digital pattern width, 0.1–1.5 mm, corresponding to the description a–k) and B) geometrical dimensions of the printed walls shown in (A) as function of the digital pattern width.
measurement using optical microscopy. The measured material volume of the printed lined was in all cases smaller than the approximated material volume. One of the reasons is the high droplet placement error and the lower energy density of the UV radiation at the high nozzle-to-substrate distance of 5 mm. Not all the printed ink is deposited at the designated position, and due to the lower intensity of the UV radiation, intense spreading of the ink can take place while the ink is still wet and not fully cured. The droplet placement error and the spreading of the ink causes a high lateral distribution of the ink forming a thin material film in the lateral range of centimeters around the initially predefined position. This thin material film is not considered in the measurement of the printed material volume and thus the measured printed material volume is smaller than the approximated calculated printed ink volume. In addition to that, material shrinking effects during UV curing as well as missing droplets during the printing process also contribute to the deviation between the material volume measured and the material volume calculated.

Based on the results obtained from the printed walls, 16 of the 3D devices were fabricated using the manufacturing technology and sequence described earlier. As an overview, some of the manufacturing steps of two of the devices are demonstrated in Figure 5. At first, the metal electrodes and the pillars were printed with the silver nanoparticle ink. The pillar diameter and height were about 200 μm and 4 mm, respectively (Figure 5A). After the electrodes, the contour of the polymer body was printed (Figure 5B) and subsequently filled by inkjet printing (Figure 5C). The size of the polymer body was about 17 × 7 × 4 mm (L × W × H). The silver pillars protrude with about 0.5 mm from the polymer body. The electrical resistivity of the printed silver pillars was measured before printing of the polymer body and after printing of the polymer body. The electrical resistivity was slightly increasing from about 0.34 ± 0.18 Ω cm to 0.48 ± 0.22 Ω cm, most probably due to a deformation of the pillar and the penetration of the polymeric material into the silver particle network. The printing time duration for one layer of the silver pillars is about 25 s. A delay time between each layer of

Figure 5. Photographs taken at different phases of the manufacturing of the printed 3D device: A) Inkjet-printed and sintered silver pillars on the printed bottom electrodes; B) inkjet-printed contour of the polymer body; C) two finished polymer bodies with the protruding printed silver pillars; D) screen-printed battery; E) final demonstrator device consisting of the printed battery and inkjet-printed silver conductors partially embedded in an inkjet-printed polymer body; F,G) are magnified images of the device showing the printed polymer bodies with the protruding pillars and the connection to the LED that is clearly G) lighting up in green color with the battery connected and F) no lighting up with the battery not connected for better visibility.
about 600 s was applied to allow the drying of each layer before the next layer is deposited on top. Printing and UV curing of two of the polymer body based on the two-step approach (printing of the contour and finally filling of the contour) takes about 240 min.

Using screen printing, a thin-film battery was manufactured (Figure 5D). Printing of the battery has been completed within 20 s. Drying and sintering time was about 10 min. The assembly of the different parts of the battery was done manually and required a time of about 3 min. The printed battery was attached to the inkjet-printed electrodes on the glass slide after the top electrodes and the LED have been deposited on the polymer block and connected to the protruding pillars (Figure 5E,F). Finally, the functional device with the green-colored LED lighting up is shown in Figure 5G. The screen-printed battery was electrically characterized. The nominal voltage was about 3 V, and the electrical charge about 20 mAh. Internal battery resistance limits the maximum current to <1 mA, decreasing with lower battery voltage. The battery was lasting more than 200 h as indicated by the LED. The LED was lighting up with very high intensity for about 5 h, intermediate intensity for the following about 100 h and finally was going off after lighting at very low intensity at about 200 h lifetime.

4. Summary and Conclusion

We demonstrated the manufacturing of a 3D electronic device combining inkjet printing and screen printing. The fabricated device has shown a permanent operation for more than 200 h. Screen printing was employed to print primary batteries used as a power source for a solid-state LED. Inkjet printing was applied for the deposition of silver electrodes and metal pillars acting as vertical interconnects on glass slides. The maximum aspect ratio of the inkjet-printed pillars was about 50:1. The inkjet-printed vertical interconnects have a height in the millimeter to centimeter range and were embedded with an inkjet-printed polymer body. A major novelty was the inkjet printing of a multi-material structure demonstrating that the material composition of 3D-printed structures can be varied. Our focus was set on the printing process and to use multiple ink droplets per printed layer, which yield to larger devices with high dimensional stability. With this combination of printing conductive and non-conductive materials utilizing inkjet technology, we could show the high potential of the manufacturing approach for many other, potentially more complex devices, e.g., in the area of microelectronics and printed circuit boards.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

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