Inkjet Printing of Electrical Connections in Electronic Packaging

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Abstract

Two aspects were evaluated of an approach to produce inkjet printed electrical connections in a roll-to-roll application. In the first part it was demonstrated that inkjet printing with silver nano-particles allows to connect LED dies embedded in a flexible polymeric substrate by way of electrical via connections and to operate the LED dies at their nominal 20 mA and 3 V driving conditions. A standard convection oven process was used to sinter the inkjet printed tracks. The second part of the work focused on identifying sinter technologies that provide the required fast processing times needed for roll-to-roll applications. IR irradiation, Rapid Electrical Sintering, and Broadband Photonic Curing were evaluated and compared with Convective Oven Sintering as benchmark technique. All these techniques produced similar track conductivities. Fastest operation was obtained with Broadband Photonic Curing, which enabled a total process time of three seconds as compared with 150 °C and 30 min in the convection oven.

Introduction

It is well known that inkjet printing provides features like additive, non-contact, digital processing. Owing to the performance of the newest generation of inkjet printheads like throughput, reliability as well as flexibility to special processing tasks inkjet printing is being introduced to the manufacturing floor.

This present work describes the integration of inkjet printing into a roll-to-roll system for the manufacture of flexible lighting devices, specifically for the printing of electrical connections to drive LED dies embedded in the polymeric substrate. Within the 'Light Rolls' EU FP7 project a novel roll-to-roll manufacturing process is developed for production of interior lighting devices based on the integration of multiple LED dies into flexible substrates. The substrate is produced in-line by way of a novel rotary RMPD® technology, self-alignment of bare LED dies, and by inkjet printing of the electrical contacts to the LEDs dies [1].

Two main aspects that were addressed separately were the inkjet printing process with nano-particle silver inks for the production of reliable electrical via connections, and the search for a fast and efficient post-treatment/sintering technique that would allow to produce low resistance electrical connections in a roll-to-roll process.

The first aspect focused on the development of the inkjet printed parameters and the specific print pattern to produce tracks on the substrate surface and electrical vias with sufficiently low resistivity to operate the LED dies. A standard convective oven process was used in this part of the work for sintering the printed silver nano-particle tracks.

Since roll-to-roll processing requires considerable feed rates, a fast sintering process is necessary to allow for compact design of the roll-to-roll machines. Several candidates for such integration were therefore evaluated in the second part of this work, with the standard convective oven sintering process being used as benchmark.

Inkjet Printing of Electrical Via Connections

Apart from the actual inkjet printing process the pre-and post-processes are of importance, and need to be developed to control the spreading of the ink on the substrate, to optimize the adhesion on the substrate, and to provide high electrical conductivity without damage of the substrates and the devices already in place.

Strong spreading of the ink can result in electrical shorts between adjacent electrical connections but may also inhibit the desired electrical characteristics, since very shallow features increase the influence of boundary scattering of electrons during conduction and, therefore, increase the resistance of the track. Several techniques can be employed to control the pattern formation on the substrate and prevent excessive spreading by either modification of surface energies [2], by mechanical constraints [3], by usage of phase change materials [4] or by application of elevated substrate temperatures in order to rapidly vaporize solvents and thereby reduce spreading.

The ink used throughout the experiments was silver nano-particle ink SunTronic™ Solsys EMD 5603 from Sun Chemical Corp, which is solvent-based and contains 20 wt% silver .

Substrates were selected to represent the final application in terms of via dimensions, wetting and thermal characteristics. Test samples were obtained from partner MicroTEC. These samples from the RMPD® process contained various combinations of via dimensions and locations as well as possible routing paths and were fabricated onto gold coated glass substrates, which acted as a common bottom electrode. The circular via opening in the RMPD® layer had a nominal diameter of 90 µm. The thickness of the RMPD® layer varied, which lead to variations in via depth of up to 10 µm at a typical average depth of 50 µm.

Printing was carried out using a binary Xaar126 printhead with a nominal drop volume of 50 pL and a maximum print frequency of 7.5 kHz with a driving voltage waveform that matched the acoustic response of the printhead to the fluidics. The printing resolution was 360 dpi, which could be achieved by tilting the printhead to an angle of 59.04° against the print direction. The meniscus pressure was held constant at -10 mbar.

Sintering in a convection oven FD (Binder, US) was studied to evaluate the onset of saturation in electrical conductivity of the printed silver nano material. For the electrical measurements, square patterns of size 20 x 20 mm^2 were printed onto polyimide foil with 3 layers of 360 x 360 dpi. The temperature of the substrate was kept at 70 °C, which controlled spreading of the ink on the substrate and facilitated drying of the silver ink.

The inkjet printed samples were exposed to 135°C, 160 °C, 175 °C, 200 °C, and 230 °C for 30 minutes and measured using a four-point probe. The results of the sheet resistance measurements indicated an exponential decrease in resistivity up to a temperature of about 185 °C beyond which no significant change with additional thermal energy could be observed. Comparison with the sheet resistance of bulk silver indicated that the inkjet printed pattern could achieve a factor of 3 of bulk resistivity of silver at oven temperatures at 185 °C for 30 minutes.

Multi-layer printing of several layers of silver ink on top of each other was shown to decrease the overall ohmic resistance due to the increase in cross-sectional area of the track. Each layer was dried at the substrate temperature of 70 °C between consecutive prints, and a single sintering step was conducted with the complete stack.

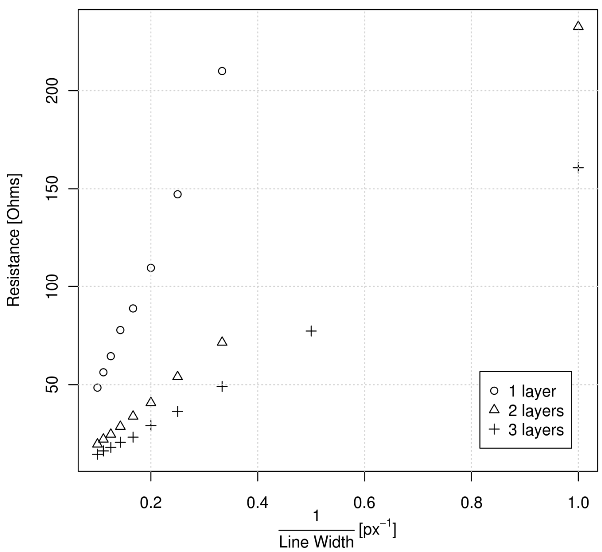

***Figure 1:** Ohmic resistance of 100 mm silver tracks versus the inverse track width*

Tracks of 100 mm length and a width in the range of 210 µm to 706 µm were produced with single, double and triple layer printing with 50 pL drop volume at 360 dpi. The substrate temperature was kept at 70 °C and a subsequent curing step of 200 °C for 30 minutes was carried out upon completion of printing of all layers.

Assuming a close-to rectangular cross-section of the printed tracks, the dependency of the resistance should be inversely proportional to the width of the track. Figure 1 clearly supports this assumption, while slight deviations from the optimum straight line of the plot indicate some deviation in line morphology.

The RMPD® process produces substrates with vias with almost vertical side walls as shown in Figure 2. The resulting sharp convex edges posed a serious challenge to the overall process, as slip was assumed to generate discontinuous metal layers or result in very thin coatings, which in turn could lead to locally high

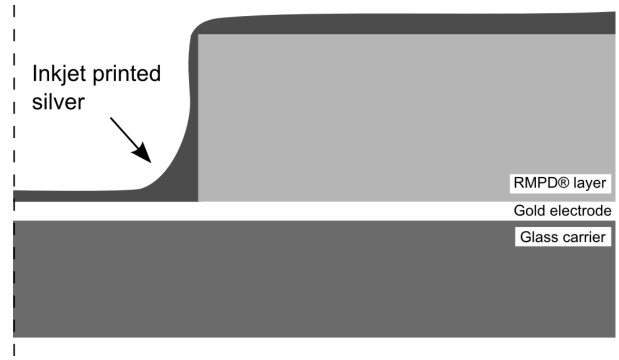

***Figure 2**: Schematic of an RMPD® layer indicating the problem to cover the sharp convex edge of the via with inkjet printing*

resistances and the tendency to burn and break the electrical connections during usage.

The studies of connecting through the circular vias in the RMPD®-layer were conducted using samples with irregularly distributed vias. Print patterns were designed that contained both, several lines of different pixel width on the top surface of the RMPD® layer, and 14 contact pads on the top surface with connection tracks to and into individual vias. The former were meant to allow for the evaluation of ohmic resistance of a connection track on the top RMPD® surface, while the latter resulted in measurement data of the ohmic resistance of the connection track, the ohmic resistance of the thin layer at the rim of the via as well as the contact resistance to the gold bottom electrode. In order to reduce effects from the top layer resistance, printed contact pads were 1.4 x 1.4 mm^2 and connection tracks of 423 µm width, much larger than the via diameter of 90 µm.

Since the LED dies to be used in the project should be kept below 150 °C for extended treatment, the process parameters for convection oven sintering were set to a temperature to 150 °C and duration of 30 minutes for the remainder of the investigation. This restricted the resistivity of the printed electrical tracks to values higher than a factor 15 of bulk silver.

Samples prepared with single layers of silver nano-particle ink showed an average resistance of 4.64 Ω with a standard deviation 2.7 Ω, while triple layers improved the resistance to an average of 1.93 Ω with a standard deviation of 0.99 Ω. The value of the standard deviation provided a measure for the reproducibility of the process and included variations of 10 µm in the thickness of the underlying RMPD® layer and thus of the depth of the via , as well as the variation in thickness of the silver deposit at the convex edge of the via.

While the variation in resistance can be partially compensated for by using appropriate circuit layouts, the long-term stability of the electrical characteristics is more critical with respect to the final application. Therefore, samples were tested with defined levels of current from 10 to 800 mA, while monitoring their electrical performance. Visual analysis was performed after the tests to identify the potential failure mechanism.

The results indicated that the generated conductive features could conduct 80 mA at 3 V driving voltage for time scales up to 60 s without change in the ohmic resistance or any visual damage. Applying the target current values, namely 20 mA for extended time and 40 mA for 600 s, showed no change in resistance and were thereby validated for the anticipated application. Currents above 100 mA resulted in varying values for ohmic resistance thus indicating excessive Joule heating of the features followed by an improvement of conductance, but also eventual disintegration. Applying currents above 400 mA lead to lift-off of the electrical tracks around the rim of the via, likely due to increased current density in the thin layers on the convex edge of the via.

In a following experiment samples with the RMPD®-layer and integrated LEDs dies were used and electrical connectors to the LED pads were inkjet printed and sintered as described. The LEDs could be successfully operated with 20 mA at 3 V, which are the nominal driving conditions of the LEDs used. Under operation the LEDs emitted bright green light as shown in Figure 3.

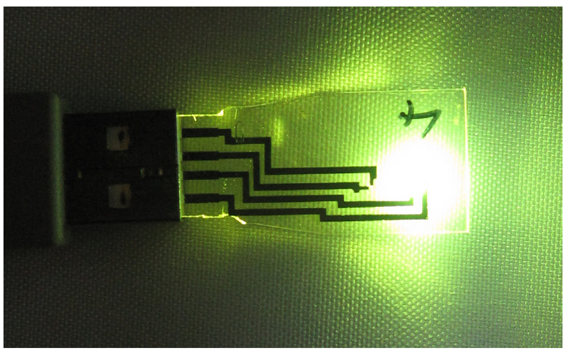

***Figure 3:** Bright green light emitted from an LED when driven with 20 mA at 3V by way of inkjet printed connections*

Sinter Technologies for Silver Nano-Particle Inks

Silver nano-particle inks contain solvents, organic binders used for stabilization, and capping polymers (e.g. poly(vinyl pyrrolidone)) on the surface of the nano-particles to prevent agglomeration. To obtain high electrical conductivity after printing a post-process is required to establish metallic contact between the silver particles by way of removing or breaking up those organic shells. The basic physics of the sintering process were reviewed by Reinhold in [5]. Many technologies have been proposed to transport sufficient energy into materials and trigger sintering, using both direct and indirect ways to elevate temperatures and, hence, allow for sintering. These techniques include classical thermal heating [6,7], laser sintering [8], microwave heating [9], exposure to UV radiation [10], IR radiation sintering [11], high temperature plasma sintering [12], field assisted sintering [13], and pulse electric current sintering [14].

Apart from thermally driven binder disintegration and diffusion the sintering processes can be based on chemical processes that break up the polymeric shells around the individual nano-particles and thus produce metallic contact between them. Several 'chemical sintering' techniques were described, which either work via touching of the printed silver nano-particle pattern with a stamp soaked in a salt solution [15,16]. Other techniques use chemicals that are either added to the fluid [17] or that are part of the coated surface [18]. These 'chemical sintering' techniques typically work fast and at low temperature, but care has to be taken to avoid undesired chemical interaction with the substrate and/or devices already present on the substrate. The present work is focused on several thermal, non-contact techniques.

A key feature for a suitable sintering technology in roll-to-roll manufacturing environments is process time, as the length of the paper/substrate path is crucial for a sensible and efficient machine design. The sintering technologies investigated in the following include convective oven sintering as a reference, infrared radiation, rapid electrical sintering (microwave) as well as broadband photonic curing.

The samples for the sintering tests were inkjet printed with silver nano-particle ink SunTronic™ Solsys EMD 5603 from Sun Chemical Corp (20%wt, solvent-based) using a binary Xaar126 printhead at 360 dpi resolution. Print pattern comprised tracks of single, double and triple layer thickness and line width of 1 to 4 pixels.

Makrofol® (Bayer, Germany) was chosen as representative non-absorbing substrate, due to its similar thermal characteristics to the RMPD®-layer as used in the Light Rolls project. The substrates were cleaned in IPA at 80 °C and the substrate temperature was adjusted between 70 °C and 100 °C to yield good spreading and line formation characteristic. The elevated temperature inhibits spreading by way of quick evaporation of parts of the solvent and thereby increasing the viscosity. This mainly controls the spreading of a deposited inkjet droplet in the initial stage of ink-substrate interactions. The temperature on top of the substrate was monitored with a TASCO IR-thermometer, focused on a blackened part of the foil surface.

Convective Oven Sintering

Convective oven sintering is a laboratory technique, which is often employed in academic research for generating conductive structures from nano-particulate deposits as it can provide a wide range of experimental settings. It is, however, a very slow process, as heat is supplied to the surface of the deposit by convection. The subsequent heat transport into the, initially, non-conductive bulk of the deposit has to be carried out by a slow, rate-limiting heat conduction process in order to allow for homogeneous temperature profiles. Especially the use of polymeric substrates with low glass transition temperatures prevents sintering at high temperatures. Though for reason of process speed and thermal limitations the convective oven sintering is not suited for roll-to-roll processing this technique was used as benchmark. Sintering was carried out at 150°C for 30 minutes using a convective oven from Binder, US.

Figure 4 summarizes the resistance per unit length measured for different layer thicknesses and widths. All experiments except for the single pixel line/single layer tracks, where line discontinuities resulted from the poor wetting of the ink on the substrate, were successful.

For the one and two-layer experiments the expected trend of linear reduction of resistance with increasing line width could be observed. As can be seen in Figure 4, strong variation of the results occurred predominantly with triple layer thickness, which may be attributed to the poor line formation as described above. The optimum condition for the combination of substrate (regarding line formation) and the applied energy available for sintering was found to be two layers and 4 pixel wide lines, yielding an average resistance of 9.8 Ω/cm with a σ of 2.2 Ω/cm, the latter resulting most probably from outliers as high as 18.3 Ω/cm. The absence of improvement with higher volume per unit loading suggests that either micro-fluidic influences, such as Taylor-type instabilities in the tracks and therefore the reduction of the minimal cross-section, or incomplete sintering by insufficient energy dose supplied for sintering.

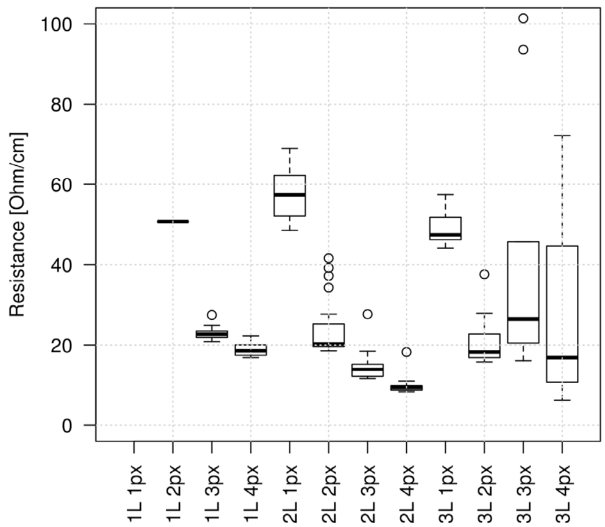

Figure 4*: Results from convective oven sintering at 130 °C for 30 minutes [Nomenclature: {number of layers} {width of line in pixel units}]*

box plot interpretation guide: thick center lines – median of the sample, box – upper and lower quartile, dashed line – minimum and maximum without outliers, single circles – outliers.

Infrared Radiation (IR)

In contrast to the above described convective oven sintering, IR radiation establishes a high temperature at the top surface of the substrate. Commercial systems typically use infrared wavelengths in the short (0.8-2 µm, filament temperature 1500-2700 °C) to medium (2-4 µm, 750-1500 °C) range, generating heat in the ink layer (short range) and heating the air above the deposits for the latter case [19]. By absorption and heat conduction, the deposit as well as the substrate heat up to around 500 °C. To prevent damage to the Makrofol® substrate the IR irradiation had to be restricted to short times, and cooling periods were introduced between consecutive IR irradiation steps.

The experimental setup used in this study was provided by Åbo Akademi University and previously used to sinter silver ink deposited by a flexographic printing process [11]. Eight sintering units (HQE 500, 500 W nominal power, Ceramicx, IRE) were placed along the paper path of the machine with intermittent spaces resulting in cycles of consecutive heating and cooling of the substrate.

The inkjet printed samples were attached to the paper web of the machine and run through the process, making sure that none of the smaller rollers (diameter 6 cm) was in contact with the top surface of the printed pattern.

The results from experiments with substrate feed rates of 5 m/min, 7.5 m/min and 10 m/min at 15 mm distance between the IR sources and the substrate, as well as one experiment at 10 m/min and 7 mm distance are presented in Figures 5 to 7.

Obtained resistances were higher than for convective oven sintering, and the data scatter was more pronounced. The latter may be attributed to different effects, such as (a) too high thermal energy at the low feed rate of 5 m/min leading to deformation of the substrate and delamination and cracking of the printed pattern, or (b) (partial) delamination of tracks as potentially caused by rapid heating of the sandwiched materials with different coefficients of thermal expansion.

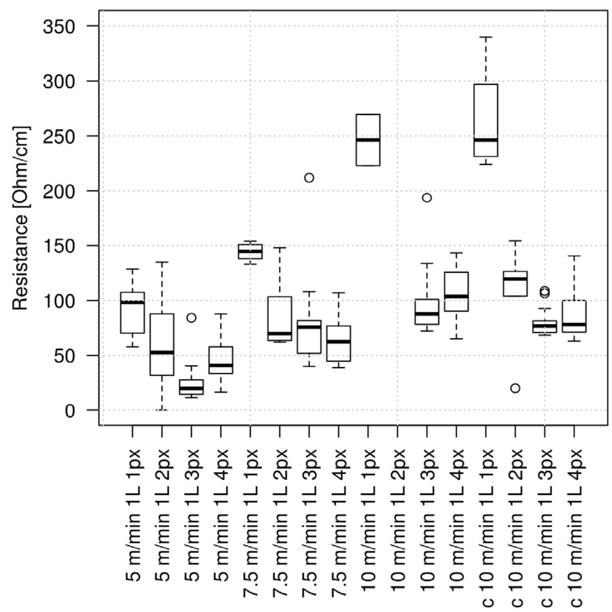

Figure 5*: Results of IR sintering of inkjet printed tracks of single layer thickness at a distance of the IR irradiator from the substrate of 15 mm. Nomenclature: {feed rate} m/min {number of layers} {width of line in pixel units} – c refers to a reduced distance between radiator and substrate to 7 mm]*

The measured ohmic resistances presented in Figure 5 to 7 indicated the superiority of double layers over single and triple layers, as data scatter was minimized and the lowest resistances per unit length were established. Taking into account the considerable bending and warping of the substrate at feed rates of 5 m/min only double and triple layers at a working distance of 15 mm were considered acceptable. IR sintering of triple layers was problematic due to local damages to the tracks potentially because of excessive heating, which resulted in the loss of several of these tracks during the test. The best results of 31 Ω/cm (1sigma of 3.8 Ω/cm) were achieved with double layers at a feed rate of 7.5 m/min and at a working distance of 15 mm.

Rapid Electrical Sintering (RES)

Rapid electrical sintering is a derivative of pulsed electric current sintering, where a current through a slightly conductive structure allows for Joule heating of the printed electrical tracks. Temperature increase is highest in the narrow contact areas

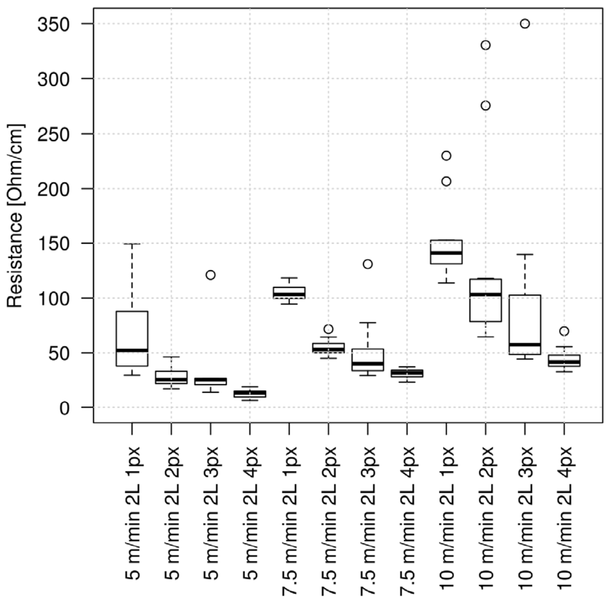

***Figure 6**: Results of IR sintering of inkjet printed tracks of double layer thickness. Distance IR irradiator to substrate was 15 mm. Nomenclature: {feed rate} m/min {number of layers} {width of line in pixel units}*

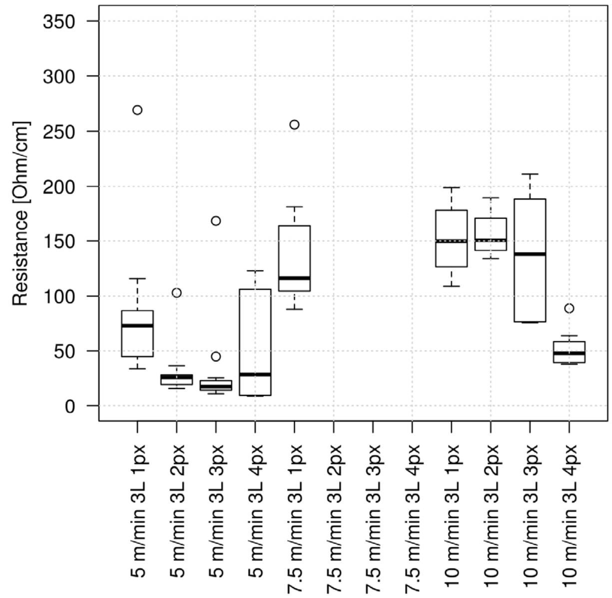

***Figure 7**: Results of IR sintering of inkjet printed tracks of triple layer thickness. Distance IR irradiator to substrate was 15 mm. Nomenclature: {feed rate} m/min {number of layers} {width of line in pixel units}*

between the individual nano-particles, which triggers the diffusion processes that lead to neck formation and sintering.

While this technique has been applied to various processes in the past, novel developments utilize rapidly changing electrical fields (radio frequency to microwave frequency) in order to accelerate electrons in the existing percolation paths. These drift currents heat up the track with the physical background described above. In this way, contactless heating of fractionally conducting features can be established. In the case of nano- particles, this process can not be applied *per se*, as the number of atoms in a nano-particle is insufficient to create fully developed conduction bands, which in turn makes the structure non-conductive.

For pattern printed with nano-particles, a pre-sintering (or annealing) step has to be conducted in order to allow for coupling of the electrical power into the structure. An alternative theory suggests that exceeding the critical field strength between particles allows for Townsend avalanche discharges, which break up the polymeric binders present in the structure and trigger initial metal-metal particle contacts and therefore create the required initial conductivity.

A problem with RES is the so-called *thermal runaway* phenomenon, as the structure becomes more absorbent the higher the conductivity and the more percolation paths are present, which will eventually overheat the sample. This problem can be approached by an adaptive RES process [20], where the reflected power from the printed pattern, which indicates the absorption rate of the incident power, is used to adjust the incident power level and thus prevents the runaway effect. This adaptation, however, is strongly dependent on the underlying amount of metallic conductors, resulting in potentially non-optimized sintering due to varying field intensities.

No adaptation of the RES process was carried out yet for the initial experiments conducted within this present investigation. Instead a generic set of RES process parameters was used at a working distance of 25µm and with reduced applied power. Nevertheless, successful sintering was achieved resulting in resistances that showed a significant drop from initially 100-150 Ω/cm after annealing to values of 10-20 Ω/cm. Further investigations with adapted RES process parameters are ongoing.

Broadband Photonic Curing

Broadband photonic curing exploits the thermal dissipation of materials during absorption of radiation, resulting in sintering and densification. Selective absorption and selected timing of the pulse train is used to design appropriate temperature ramps for fast sintering of the absorbing material. Commercial systems have lately entered the market for printed electronics due to their potential for roll-to-roll processing of conductive pattern. Feed rates are proposed up to 60 m/min. Emission spectra range from 220 nm to 1000 nm and can be pulsed at frequencies greater than 1 kHz [21].

A set of samples as described above was sent to Novacentrix (Austin, TX) and treated in a Pulseforge 3100 tool with an optimized set of process parameters, that were assessed by Novacentrix in preceding experiments for this specific combination of substrate and silver nano-particle ink. The feed rate used in this experiment was 10 m/min, and with the length of the curing lamp assembly of 0.5 m the interaction time of the single pass treatment resulted in approximately 3 seconds.

The compiled results in Figure 8 show low data scatter for both the double and triple layer tracks. Interestingly, double and triple layers produced similar conductivities, which may be due to line formation characteristics at higher volume loading as insufficient drying of the triple layer features allows for dynamic changes of the track morphology. Minimum resistances were found to be 9.6 Ω/cm (1sigma of 2.3 Ω/cm), which is essentially identical to the results from convective oven sintering (9.8 ± 2.18 Ω/cm). It has to be kept in mind that Broadband Photonic Curing requires pre-dried samples, which could be provided e.g. by a low power IR lamp.

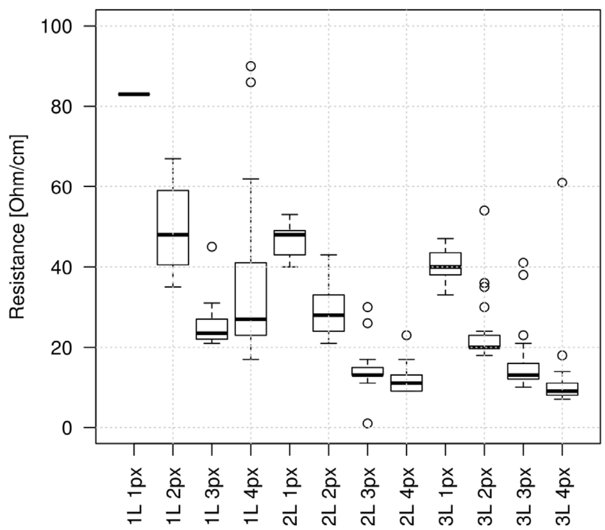

***Figure 8:** Results from Photonic Sintering using optimal conditions determined by Novacentrix [Feed rate: 10m/min, processing length: ~50 cm, Nomenclature: {number of layers} l {width of line}px]*

Conclusion And Outlook

Table 1 compares the sintering results and summarizes information about the different sinter techniques investigated. All techniques enabled sintering of tracks of inkjet printed silver nano-particles, and to produce resistivity data similar to that of convection oven sintering, which served as reference.

IR irradiators are readily available and can be easily integrated. The necessity of cooling stations/chucks has to be investigated in the running system. The cooling cycles that are necessary between the irradiation periods require chucks and require time, which increases the total cycle time of the sinter process and thus the total length of the roll-to-roll work station.

Rapid Electrical Sintering RES appears as a very elegant solution for establishing a hybrid sintering process for single pass processing at a path length below one meter. It remains to be proven, however, that it can be adapted to generic pattern, and that it can be integrated in roll-to-roll machines. No commercial systems are available yet. A further concern is the necessity of pre-sintering of the samples because RES requires a certain minimal conductivity of the pattern to initiate the sintering process.

***Table 1:** Overview of the investigated sintering technologies with respect to their achieved resistance per unit length, thermal impact onto the substrate, ease of integration and approximate length of the sintering path*

	Min. Achieved Resistance [Ω/cm]	Thermal Impact on Substrate	Ease of Integra-tion	Approx. Length of Curing Path @ 10 m/min
Convective Oven	10	High	Easy	300
Infrared Radiation	25	High	Easy	~5-10
Rapid Electric Sintering	10-15	Medium/High	Difficult	< 1m
Photonic Curing	10	Low	Easy	< 1 m

Broadband Photonic Curing produced the essentially the same electrical conductivity as convective oven sintering, however, at a total process time of three seconds, which allows for highly compact assemblies in roll-to-roll machines. Some pre-drying of the sample is required. Commercial systems are available, but at considerable cost. This promotes photonic sintering as a potent candidate for roll-to-roll applications.

As was demonstrated in the first part of this work it was possible to electrically connect LED dies embedded in polymeric RMPD® layers by inkjet printing of electrical tracks and through electrical via connections, and thus allowing for operation of these LEDs at 20 mA and 3 V.

Based on the presented results in this work focus will be laid on implementing Broadband Photonic Curing into the RMPD roll-to-roll application within the Light Rolls FP7 project. Next steps will be the sintering of RMPD samples with inkjet printed circuitry and embedded LED dies.

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Biography

Ingo Reinhold graduated in micromechanics-mechatronics with emphasis on print- and media technology from Chemnitz University of Technology in 2008. After joining Xaar’s Advanced Application Technology group in Järfälla, Sweden, he focused on advanced acoustic driving of piezo-type inkjet printheads alongside with pre- and post-processing of functional materials in digital fabrication. He is currently enrolled as a PhD student within the iPack VINN Excellence Center at the Royal Institute of Technology (KTH) in Stockholm, Sweden.