Electrophotographic Printing of RFID Antenna Coils on Cofired and Postfired Ceramics

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Abstract—Digitally printed electronics are a major field of activity in research and industrial applications nowadays, especially in thick-film technology. Within the last decade great progress has been made in print electronics toward the goal of supplementing or replacing the established screen printing technology. The advantage of digital printing is its capability for rapid production without any layout-specific tools (i.e., screens). This highly flexible production method is especially applicable for prototyping and small series productions.

Electrophotography, commonly known as laser printing, has been considered very rarely as a method for digital printing of electronic circuits. It is a completely solvent-free method with high speeds and a high potential for precision. At Helmut Schmidt University, electrophotography is being developed as an alternative method to print silver lines as a basis for a conductive layout.

Following the publication of the first set of conductive silver lines to be printed, this paper covers the development from initial conductivity to a functional circuit element. Deficits in toner transfer to tape are examined and a more effective transfer method is established. Furthermore, the used silver toner is improved, and its performance is tested regarding the desired application. Silver lines are printed on green tape and sintered in a cofiring process, as well as on ceramic using a postfiring process. After reaching a certain level of quality, a set of functional RFID antenna coils is printed that proves the capability of the process to create a conductive layout.

 $\it Keywords$ —Electrophotography, conductive silver lines, printed electronics, RFID

Introduction

Digital printing of electronic circuits is an essential issue for many current applications and research works. In recent decades, great progress has been made in implementing digital methods in order to supplement or replace established screen printing.

At the Institute of Automation Technology of Helmut Schmidt University (HSU) in Hamburg, Germany, electrophotography, commonly known as laser printing, is investigated as a production method for printing conductive layouts for thickfilm applications.

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The electrophotographic process, shown in Fig. 1, consists of several stages that exist in different variations. In the following, the process is described as it is used in the present study.

Its core is an organic photoconductor drum (OPC) that rotates inside the printing device. Initially, it is charged via a corona. An LED bar selectively discharges spots on the OPC so that a latent image of the desired picture is formed. The drum passes the developer station, where toner particles adhere to the discharged areas of the OPC.

Before the toner can adhere to the drum, they must have charge attached, which is accomplished by tribocharging. Inside the developer station, a two component mixture of toner and ferrite carrier particles are stirred mechanically. Thereupon, the carrier and toner particles tribocharge each other. The properly charged toner particles are developed onto the OPC while the ferrite carrier particles remain inside the developer station.

Afterward, the toner particles are transferred to the substrate, where they are fused using an IR heater. The OPC is cleaned and discharged and the process starts again [1, 2].

Electrophotography is a very common method for conventional printing applications like office printers, book printing, and so on. However, up to the current time, very few approaches were published to use it as a method for printing functional particles. Experiments were made to test conductive copper powder in an electrophotographic environment [3]; also printing of 3D objects has been investigated [4]. A patent exists regarding electrophotographic printing of circuit patterns or even of multilayer wiring boards [5], but so far no published results of printing a conductive layout are known to the authors.

In cooperation with CtG PrintTEC and Zobrist Engineering, silver toners are tested in a printer prototype, with the goal of printing conductive silver lines as a basis for electronic circuitry.

The feasibility of the technology was generally proven and formerly published [6], but the results left room for further improvement.

In this paper, the printing device is improved and new toner is implemented. This paper describes the progress from initially printing small amounts of silver on green tape to functional laser-printed conductive elements, specifically RFID coils, on ceramics.

A. Toner Transfer to Substrate

The transfer of toner to the substrate is one of the stages in the electrophotographic process. Usually, in graphic

applications, carbon black or colored toner is transferred from the OPC to paper.

Two common methods exist: A transfer roller or a transfer corotron [1]. In its first version, our printer was equipped with a transfer roller, where the substrate is guided in between the OPC and the roller. A transfer voltage is applied to the roller. Thus, the toner particles are transferred via mechanical as well as electrical force from the OPC to the substrate. This method has two major disadvantages. First, it is only possible to print onto elastic substrates, which excludes already fired ceramics. Second, research results show that the toner is not transferred well onto green tape [6].

Usually, with this transfer method, the toner transfer saturates at a certain transfer voltage [7]. However, contrary to printing on paper, the saturation level is not reached when printing on green tape, as Fig. 2 shows. Therefore, a high amount of toner stays on the OPC and is not transferred onto

To fusing station Waste toner Erasure Transfer station AC-corona Cleaning unit Paper path • Discharge lamp Transfer corotron Photoconducter drum Charging corotron Developer Station LED print bar Control Potential sensor

Fig. 1. Schematic overview of the electrophotographic process [1].

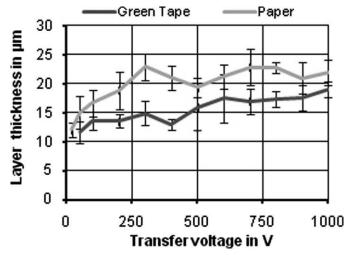


Fig. 2. Deposition of carbon black toner on paper and green tape with a conventional transfer roller (bars represent standard deviation).

the tape. Further experiments confirmed that this effect occurs with silver toner as well.

These results led to a redesign of the printer. As a change of minor importance, a so-called conditioning roller was removed from the printer. This roller was a patented feature of the first printer prototype [8]. It was used in the preliminary study [6], but it never had a proven effect.

More importantly, the transfer method was changed. Instead of using a regular transfer roller as mentioned above, the toner was transferred from the OPC onto a special transfer roller, from which it was deposited on the substrate, as Fig. 3 shows. The substrate was charged via two coronas. This patented method is only applied in a few prototypes; it is uncommon in regular electrophotographic applications [9].

Still, toner is transferred via mechanical as well as electrical force. However, with this method, it can be printed on inelastic substrates, such as fired ceramics. Furthermore, the gap between the transfer to paper and the transfer to green tape is closed. Fig. 4 shows the results of an experiment where carbon

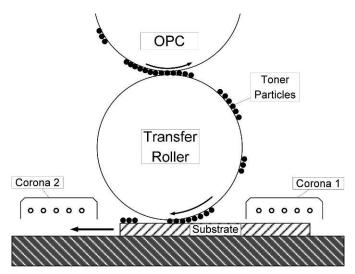


Fig. 3. Toner transfer via surface charge as used in the printer prototype at HSU.

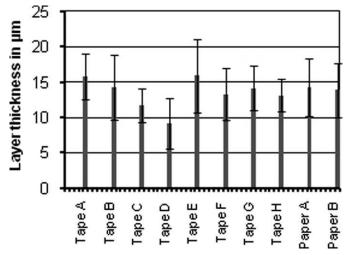


Fig. 4. Deposition of carbon black toner on different green tapes and papers (bars represent standard deviation).

black toner was printed on eight different types of green tape as well as two types of paper.

No significant differences in the deposition of these substrates were observed, except for tape D. Test prints with silver toner show a more homogenous transfer of toner on green tape than before, as evaluation of the optical line density shows.

However, layer thickness does not reach the maximum level achieved with the former transfer method. It could be that a high amount of toner stays on the transfer roller and is not transferred to the substrate. Under this assumption, the surface properties of the roller are responsible for this drawback. However, it also has to be considered as a general drawback of this method.

As a positive result, the surface charge via the coronas works very well with green tape. A solution using direct deposit of the toner from the OPC toward the substrate (without the transfer roller) should be considered and examined.

B. Toner Improvement

In addition to redesigning the printer, further development of the silver toner was necessary. The main issue when creating a toner from conductive silver powder is ironically its conductivity. A toner must be nonconductive in the electrophotographic process. Otherwise, it would lose its charge quickly and also cause short circuits inside the printer.

Zobrist Engineering from Switzerland has the capability of producing silver toner. However, this toner comes with several limitations. It is not as chargeable as a conventional toner, and too many particles with the wrong charge polarity occur. Furthermore, it is only possible to use spherical silver particles as a basis; a toner based on flakes has not worked so far. Nevertheless, as already mentioned, the feasibility of the process could be proven with this first generation toner, called C01.

To overcome these limitations, a method using pretreated silver particles is implemented at HSU. As a first test, a mixture of spherical and flake-shaped silver particles was pretreated, and a toner called C02, containing 69 wt% of these particles, was manufactured by Zobrist Engineering. Fig. 5

shows its charge distribution, measured with the EPPING q/d-meter [10].

It is remarkable that it is possible at all to create a toner out of these particles. Prior tests to do so failed. The toner shows an acceptable charge, high homogeneity, and a relatively low number of particles with the wrong polarity.

In addition, another toner called C03 is produced. It is based only on spherical silver particles, which were equally pretreated and afterward processed into a toner. Compared to C02, it contains an equivalent amount (69 wt%) of silver and promises even better charge properties. Both toners are tested in this study for printing conductive layouts. With these toners, it is possible to print very homogeneously on green tape and to generate silver toner lines with a higher density than before when performing only a single printing cycle. Such a line is displayed in Fig. 6.

Still, particles are deposited around the line where they are not intended to be. Although the density of the lines could be improved as shown in prior tests [6], a single printing cycle still is not sufficient to generate a conductive line after firing.

Thus, multiple print cycles have to be considered as a solution to create conductive silver lines. To do so, it has to be determined whether the toner particles are fused between the print cycles or not. It could be that the charge of the transfer roller removes toner particles from the substrate with further print cycles. However, as Fig. 7 shows, the amount of toner deposited on the substrate stays in general pretty equal with every additional print cycle, with fusing as well as without.

Therefore, both methods can be applied in the process. However, if inaccuracies in positioning occur, fusing in between cycles prevents toner from getting removed from the substrate due to the charge of the transfer roller.

C. Cofiring Process

To receive a functional layout after printing, a firing process was necessary. For this purpose, silver lines were printed using C02 toner with five and 10 printing cycles onto Ceramtec LFC100-250CT green tape. Afterward, the tape was cut into squares with a width of 2 in. Four layers of tape were laminated with a pressure of 200 kN. Only cold lamination was

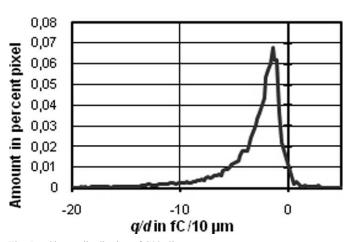


Fig. 5. Charge distribution of C02 silver toner.

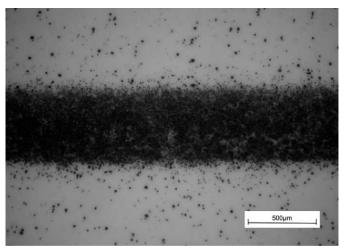


Fig. 6. Printed line of silver C02 toner (before firing).

possible, because the toner particles melted slightly otherwise and adhered to the press. Release tape was not used to cover the tape because firing experiments showed that the adhesion between this tape and the silver lines was so strong after firing that it could be removed.

In a first approach, the laminated green tapes were fired with a heating rate of 5°C per minute to a maximum temperature of 875°C with a dwell time of 15 min before cooling down with equal rate. Although different firing profiles (maximum temperature, heating rate, dwell time, steps in between) were applied, the effects described in the following still occurred.

Without release tape, the ceramics shrank about 20% in side length which equals approximately 36% in area. Additionally, the ceramic bent significantly, as Fig. 8 shows.

This bending effect seems to be caused by a differential shrinking behavior of tape and toner, based on the silver particles used. The bending can be easily neutralized by using equally printed tapes on both sides of the ceramic. In this case the ceramic was flat after firing. This is not a very satisfying solution, because mechanical tension can be expected in the

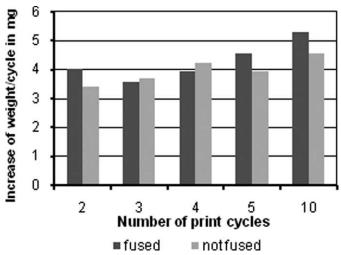


Fig. 7. Toner transfer with a defined image at multiple print cycles (silver toner).



Fig. 8. Bending effect of C02 silver lines on a cofired ceramic (2 in \times 2 in).

surface area. However, it is considered sufficient for further testing.

A more severe effect is the appearance of large traverse cracks within the generally well shaped, but also edgy silver lines, as shown in Fig. 9. The lines have a relatively high density and also conductivity can be measured on short tracks.

The cracks occur in a large number on short sections. The already mentioned variation of firing profiles does not yet lead to any satisfying results. Neither slower increase of temperature, nor burnout at different stages, nor additional firing at above 900°C have so far eased the problem.

Another experiment to suppress the appearance of these cracks was to spin-coat the tape with a glass paste above the silver lines. However, although the coating worked very well and a fair layer of glass was built up above the lines, the cracks still appeared beneath the glass as seen in Fig. 10.

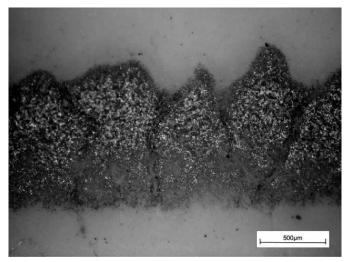


Fig. 9. Traverse cracks in silver lines after cofiring process.

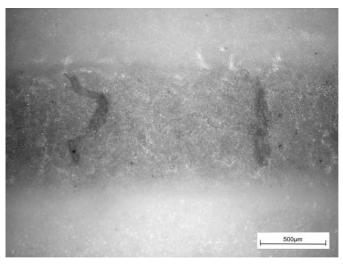


Fig. 10. Traverse cracks in silver lines beneath a glass layer after cofiring process.

Obviously, it is not possible to overcome this challenging aspect with C02 toner. The toner adhesion to the substrate is basically controlled via the amount and properties of the glass that is added to the toner. This would seem to have a big impact on the occurrence of cracks.

Glass is added to the toners used in order to achieve ceramic surface adhesion. In C02, the glass is not optimized regarding the chosen green tape. This was the idea behind the C03 toner design, which had a higher amount of glass. Furthermore, the glass is adapted to the green tape composition.

C03 toner performs very well on green tape and in the sintering process. Fig. 11 shows a cofired silver line, fired with a heating rate of 5°C per minute via a 2 h burnout at 300°C to a maximum temperature of 875°C with a dwell time of 15 min before cooling at an equal rate.

The cracks observed with C02 do not appear, and a very homogeneous and sharp silver line with a high density is formed after firing. Still, some undesired effects occur. The bending of the ceramic has not been solved in a satisfying manner, but again it can be neutralized by printing on both sides. Actually, the problem even intensified due to the good performance of the toner. It adheres so well to the substrate that a too high amount of toner creates such a force, that several substrates broke. The toner must therefore be further adapted to the substrate.

Nevertheless, very well shaped cofired silver lines can be created with a low failure rate and high conductivity. Sheet resistance of these lines ranges between 3 and 15 m Ω , depending on the geometry and number of printing cycles. Thus, these printed silver lines yield a good base for the desired cofired RFID coils.

D. Postfiring Process

In addition to exploring the possibility of cofiring laser printed silver lines and therefore RFID coils, a postfiring process is also considered. For this purpose, silver toner is printed on an already fired aluminum oxide ceramic. This is feasible due to the changes in transfer described earlier in this paper.

500µт

Fig. 11. Cofired silver line, printed with C03 toner.

Challenges occurred with printing the first generation toner, C01, onto the ceramic, as Fig. 12 shows.

The toner does not transfer very well and 10 printing cycles were performed to get an acceptable amount of toner onto the substrate. After firing at 650°C (5°C heating per minute, 1 h dwell, 5°C per minute cool down), a silver line with a very low density formed, so that only very little conductivity on inacceptably short tracks was achieved.

The improved C02 toner shows significantly better performance. After five printing cycles, the amount of toner on the substrate was considered sufficient. Problems occur with the above mentioned removal of formerly deposited toner by the transfer roller, due to inaccuracies in hitting the lines with further printing cycles. This can be solved by fusing the toner in between cycles. However, the inaccuracies with the current printer setup lead to broader and less sharp lines than desired.

Nevertheless, relatively well shaped silver lines, as displayed in Fig. 13, are produced after firing at 650°C (5°C heating per minute, 1 h dwell, 5°C per minute cool down).

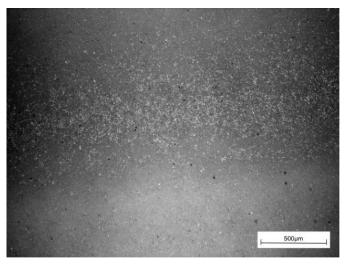


Fig. 12. Laser printed silver lines on ceramic using first generation C01 toner (postfiring process).

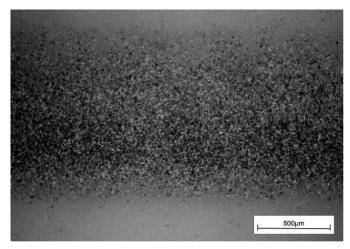


Fig. 13. Laser printed conductive silver lines on ceramic using second generation C02 toner (postfiring process).

All printed lines are conductive over their full length (96 mm) without interruptions. Lines with a width of 1 mm show a resistance of 8-11 Ω , and lines with a width of 0.5 mm show a resistance of 17-24 Ω , both with very little deviation from measured values (<0.5 Ω).

Even better results can be achieved with the C03 toner. Fig. 14 shows a silver line on ceramic after a postfiring process, applying a heating rate of 5°C per minute via a 2 h burnout at 300°C to a maximum temperature of 875°C with a dwell time of 15 min before cooling down with the same rate.

The minimum realized line width is about 300 μ m with a sheet resistance in the range of 25 m Ω (three printing cycles), and a sheet resistance of 2 m Ω can be achieved at a 1.0 mm line (10 printing cycles) [11].

Further research is necessary to determine which factor is decisive regarding conductivity and performance. While the amount and choice of glass seems to have a massive impact on sintering the silver lines in the cofiring process, it could not yet be determined which of the differing factors (firing profile, particle shape, glass) is definitive regarding conductivity in the postfiring process. A larger amount of toner first has to be produced and tested to give clear answers to this question.

However, compared with the cofiring process, the generation of conductive silver lines using the postfiring process works pretty easily and without major drawbacks. The fact that the ceramic does not shrink during the process seems to improve the adhesion between silver particles and substrate. Cracks do not occur with both toners and mechanical force does not seem to have any impact on the lines and the ceramic.

E. Printing RFID Antenna Coils

An antenna is a conductive structure, specifically designed to couple or radiate electromagnetic energy. An RFID antenna, in certain ranges, is composed of a coil (inductive loops) to which the RFID chip gets attached [12].

Using the knowledge described in the prior sections, only a small step was necessary to produce an RFID antenna coil.

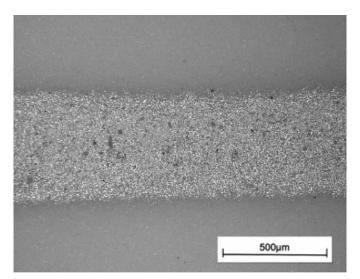


Fig. 14. Laser printed conductive silver lines on ceramic (C03 toner, postfiring process).

Two simple layouts are chosen to basically represent such antennas, a single looped coil with a diameter of approximately 48 mm and a line width of 2 mm, and a triple looped coil with equal diameter (outer loop) and width. The chosen width avoids problems with inaccuracies in multiple printing. First, C02 toner was tested in printing such coils. Five printing cycles were performed on already fired aluminum oxide ceramic and the toner was fused in between. Pictures of the printed coils after firing at 650°C (5°C heating per minute, 1 h dwell, 5°C per minute cool down) are displayed in Fig. 15.

The single looped coil has a resistance of 5.6 Ω and an inductivity of 0.3 μ H while the triple looped coil's resistance is 19.2 Ω with an inductivity of 0.8 μ H.

Even better results can be achieved when using C03 toner. The above mentioned layout is used again, but complemented by two more sets of coils with line widths of 1.0 mm and 0.5 mm. A picture of such printed coils is displayed in Fig. 16.

The coils were sintered in a postfiring process, with a heating rate of 5°C per minute via a 2 h burnout at 300°C to a maximum temperature of 875°C with a dwell time of 15 min before cooling down at the same rate. The printing and firing process worked without any problems and no failures in printed coils were detected. The properties of the coils were measured using an LCR meter, considering the coil as a series connection of resistor and inductor. The measurements show stable values with only little variation. The results are presented in Table I.

Assuming a typical RFID frequency f of 915 MHz, the quality factor

$$Q = \frac{2\pi f \cdot L}{R}$$

of these coils is in range of 370-2400, which is far better than required.

Besides the good results in the postfiring process, the improved performance of the C03 toner also gives the opportunity of printing cofired RFID coils. To do so, four layers of C03 toner were printed on green tape, and the tapes were sintered with the same firing parameters as already used in the postfiring process. The respective coils are shown in Fig. 17.

Printing and cofiring the coils is slightly more challenging than postfiring them. Due to the high amount of silver, the thicker coils (2 mm width) break occasionally during the firing process and a certain failure rate occurs. Furthermore,

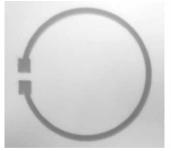




Fig. 15. Laser printed postfired RFID coils (C02 toner) with an outer diameter of 48 mm, and a width of 2 mm.

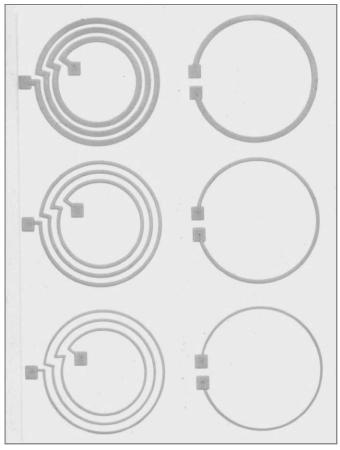


Fig. 16. Laser printed postfired RFID coils (C03 toner) with an outer diameter of 48 mm and widths of 2.0, 1.0, and 0.5 mm.

Table I
Resistance and Inductivity of RFID Coils (C03 Toner, Postfired, Four Printing Cycles)

| Number of loops | Line width (mm) | L (µH) | R (Ω) |
|-----------------|-----------------|--------|-------|
| 3 | 2.0 | 0.72 | 1.74 |
| 3 | 1.0 | 0.78 | 3.46 |
| 3 | 0.5 | 0.86 | 8.03 |
| 1 | 2.0 | 0.31 | 0.86 |
| 1 | 1.0 | 0.34 | 1.71 |
| 1 | 0.5 | 0.40 | 6.15 |
| | | | |

the bending effect has a much bigger influence because it is not that easy to exactly position the same layout on the backside to overcome the effect. However, Table II shows some results of inductivity and resistance of cofired RFID coils (also considered as a series connection).

Compared with the values of postfired coils, the variation is stronger and, due to the failure rate, fewer coils can be examined. Taking these problems into consideration, nevertheless, Table II gives a cautious impression of the properties of the successfully printed cofired RFID coils.

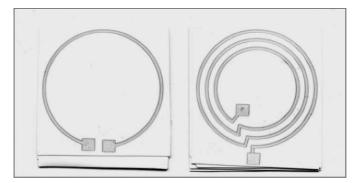


Fig. 17. Laser printed cofired RFID coils (C03 toner) with an outer diameter of 48 mm, and a width of 1.0 mm.

Table II
Resistance and Inductivity of RFID Coils (C03 Toner, Cofired, Four
Printing Cycles)

| Number of loops | Line width (mm) | $L \; (\mu H)$ | $R(\Omega)$ |
|-----------------|-----------------|----------------|-------------|
| 3 | 2.0 | 0.63 | 1.08 |
| 3 | 1.0 | 0.67 | 2.24 |
| 3 | 0.5 | 0.68 | 4.84 |
| 1 | 2.0 | 0.42 | 0.42 |
| 1 | 1.0 | 0.28 | 1.52 |
| 1 | 0.5 | 0.30 | 4.05 |

Conclusions

The experiments performed at HSU show that conductive silver lines, and therefore RFID antennas, can be printed using electrophotography. The main advantages of this method are the speed of the process and the high flexibility, allowing for the possibility to print without any additional tools such as a screen. Any layout desired could be printed without modification of the printer.

The results of printing on green tape are improved significantly by changing the method of transfer and by developing an improved toner. The toner allows for printing on green tape and sintering the coils in a cofiring process. However, it also yields new challenges for future research. The results are not yet sufficient, especially concerning the remanent tension, bending, and breaking of the ceramic, show that further adaption of toner and tape is necessary. And, although the change of amount and type of glass seem to be a decisive factor, the influence of other factors, such as the firing profile and the silver particles used, must be examined in more detail in the near future.

Regarding the postfiring process, both toners examined work very well in printing silver lines as well as RFID coils. Well-shaped coils with satisfying electrical properties could be printed, but further research is necessary to learn more about the influence of the firing process and the particles used.

In addition to the ceramic base used here, it is also within the realm of possibility to print on many other substrates. For example, printing an antenna on product packaging together with graphic toner might be a large field for this application. The biggest challenge will be the development of a method to achieve conductivity without a sintering process. Furthermore, printer development has to be pushed further. This is a critical challenge due to the costs that apply. However, progress in toner performance, especially regarding C03, is so huge that the printer reaches its constructional limits. It has to be pushed further in order to print smaller structures with better scalability.

Nevertheless, the results show the huge potential of electrophotography for manufacturing conductive layouts of many kinds. Research at HSU will continue to explore the possibilities in printed electronics.

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