

# Roll-to-roll infrared sintering of gravure printed silver patterns in applications of back-injection-molded functional lightweight structures

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## Abstract

We report on the manufacturing of functional lightweight structures by the combination of roll-to-roll gravure printing technology and back injection molding technology. The objective is to manufacture conductive grid patterns consisting of thin lines in the lower micrometer range integrated in lightweight components. We already reported on the roll-to-roll manufacture of thin conductive lines by gravure printing of a nano-silver ink which is rapidly sintered by infrared radiation at web velocities up to 1 m/s [1][2]. This approach is employed for the fast manufacturing of conductive grid patterns on a flexible substrate. Afterwards, the printed patterns are integrated in lightweight structures by back injection molding. The final composites shall be applied as smart components for example in cars and rotor blades of wind turbines for electrical applications like power supply, electrical circuits and data communication [3].

## Introduction

The reducing of time and production costs as well as a resource efficient manufacturing are important aspects in every production field. The printing of thin lines in the micro scale on flexible substrates by roll-to-roll gravure printing and its integration into lightweight structures by injection molding has great potential to fulfill the mentioned requirements. The roll-to-roll printing contributes to a time saving production and the possibility of the integration into a mass manufacturing process.

After printing, the patterns get dried and sintered in-line. This process lasts less than two seconds.

The printed thin lines should contribute to the realization of semi-transparent grid electrodes for power supply, as electrical circuit or in communication applications. With the grid structure introduced in this paper the electromagnetic radiation of sensors could be influenced or the efficiency of antennas could be improved without the use of additional electronic devices. They could also be applied in touch sensing elements or in stress sensors of car and rotor blade components [1].

To realize a sufficient performance for the particular applications certain parameters have to be taken into account accurately. The parameters include the grid constant and line width of the printed grids as well as the thickness and type of printing substrate and the injection molding material.

## Experimental setup

### Process Development

In comparison to other approaches [4][5] a high-throughput method to print, dry and sinter a conductive grid pattern on a flexible, low-cost substrate is presented.

The process steps to manufacture roll-to-roll gravure printed silver patterns which are cured by infrared sintering and applied into back-injection-molded functional lightweight structures are as follows:

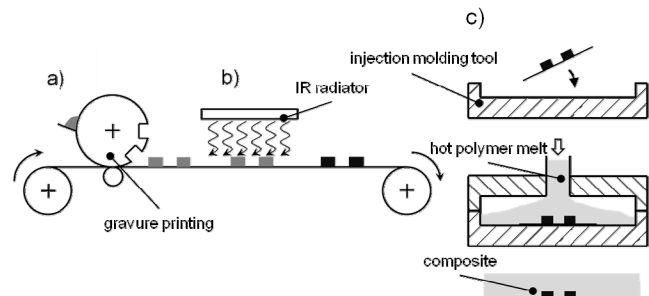


Figure 1: Manufacturing of functional lightweight structures by R2R gravure printing and back injection molding. a) R2R gravure printing of grid pattern, b) R2R IR drying and sintering, c) back injection molding

- (1) Development of a gravure printing form to print grid pattern with a line width below 100  $\mu\text{m}$  and line distances less than 5 mm.
- (2) The silver nano-particle ink (PFI-722 from PChem Associates, Inc.) is rotogravure-printed (figure 1, a) on a moving web of PET foil (Melinex 401CW from DuPont Teijin Films).
- (3) Drying and sintering is done in-line within less than 2 seconds (figure 1, b). Therefore, the applied infrared module (Heraeus Noblelight) has a length of 500 mm.
- (4) The grid patterns were cut from the web and processed by back injection molding (figure 1, c). Here, known mass production polymers are used.
- (5) The composites are optically and electrically characterized to figure out the effect of back injection molding regarding the geometrically grid stability as well as the impact on the electrical properties.

## Machinery

The printing experiments are done by the hybrid roll-to-roll laboratory gravure printing machine by LABORMAN (manroland). The machinery layout is shown in figure 2.



Figure 2: Roll-to-roll rotogravure printing machine LABORMAN from formerly manroland.

It has a maximum web width of 140 mm and a maximum web velocity of 60 m/s. The equipment of the printing machine consists of a gravure printing unit using sleeve technology combined with an inking unit and a doctor blade [1].

The layout of the printing form (see figure 3) is designed digitally and produced by the gravure sleeve manufacturer Sächsische Walzengravur GmbH (SWG) using direct laser engraving. With this process Raster Image Processing (RIP) could be avoided, every pixel of the pattern geometry could be manipulated and continuous lines can be realized. The laser beam shot has a diameter of 20  $\mu\text{m}$  which has to be considered in the pixel arrangement.

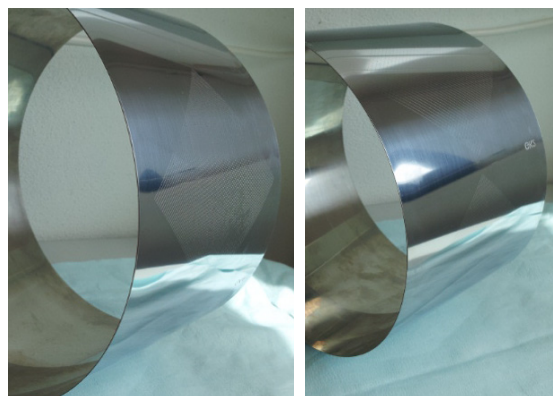


Figure 3: Gravure printing form with engraved grid patterns.

The printing system is completed by an IR dryer with an electrical power supply of 35 kW [1].

Injection molding was done using a KraussMaffei KM-250-1400-C2 machine (KraussMaffei Technologies GmbH, Munich, Germany) to produce injected plates with dimensions of 150x250x4 mm.

## Materials

To realize sufficient electrical conductivities metal inks are applied. They are mostly solvent or dispersion based and they consist of small metal particles in micro down to nanometer ranges. Because of the realization of thin lines an ink with spherical particles is used. Literature shows that especially silver nano-particle inks are characterized by a good printability and an excellent conductivity after sintering process [1]. In printing specific rheological requirements of the inks have to be taken into account. In gravure printing the ink should have a viscosity between 10 and 200 mPas [6].

The ink which is used to produce thin lines in gravure printing is PFI-722 by PChem Association Inc., which is an aqueous dispersion with an average particle size of 10 to 30 nm. Its silver content is 60 wt% and the viscosity represents 130 mPas.

The structures are printed on the PET substrate Melinex 401 CW by DuPont Teijin Films with a thickness of 100  $\mu\text{m}$ , no acrylic coating and one-sided slip pretreatment [1].

The back injection polymer used for the neat preparation is thermoplastic polyurethane (TPU) (Desmopan 460, Bayer Material Science AG, Leverkusen, Germany).

## Results and Discussion

### Gravure printed grids

As already mentioned the patterns for the gravure printed grids are generated digitally with a resolution (pixel distance) of 5  $\mu\text{m}$ . Preliminary experiments were already determined to realize finest line widths. The experiments show that the alignment of 3 pixels in an angle of 45° results in line widths in the range of 40 to 55  $\mu\text{m}$  based on the mentioned laser beam shot diameter of 20  $\mu\text{m}$ .

Different grid patterns with five grid constants are generated. Figure 4 shows the unit cells of the single grid constants from grid constant (GC) 1 to GC5. In figure 4 (f) an enhanced grid line of the digital pattern could be seen. The characteristics of all grids are summed up in table 1.

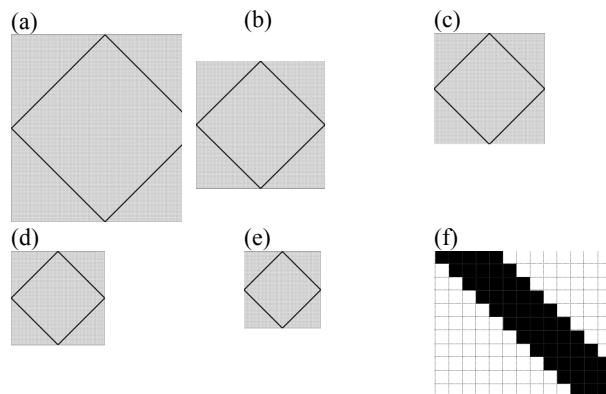


Figure 4: Digital pattern of the grid unit cells. (a) GC (grid constant) 1, (b) GC2, (c) GC3, (d) GC4, (e) GC 5, (f) Enhanced grid line

	Theoretical grid constant [mm]	Resolution/ Pixel distance [mm]	Angle [°]	Line width [px]
GC1	2.2	0.005	45	3
GC2	1.5			
GC3	1.3			
GC4	1.1			
GC5	0.9			

Table 1: Parameters of the digitally generated grids.

The line widths of the grids are uniform, only the grid constant (GC) is changed as it can be seen in table 1.

The aim of the grid generation with different grid constants is the realization of semi-transparent grid electrodes as already mentioned in the introduction. The grids are applied in large-scale (80x80 mm) onto the printing form.

In figure 5 the comparison between engraved lines on the sleeve and a printed line on substrate is shown. Figure 5 (c) depicts the microscopic image of GC5 with a grid constant of 0.9 mm which is printed with a printing speed of 0.5 m/s.

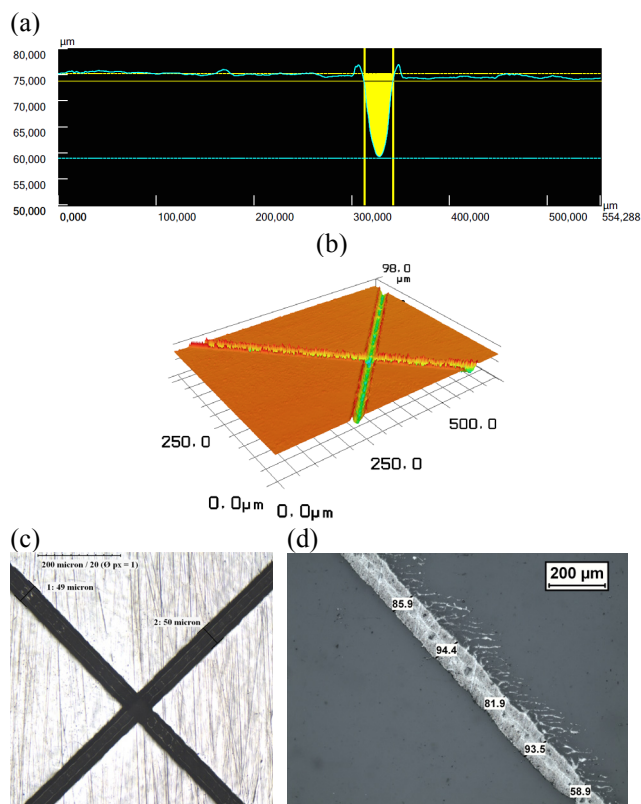


Figure 5: Comparison of printing form design and printed structures. (a) Channel profile of line engraved on printing cylinder, (b) 3D channel geometry of engraved lines, (c) 2D microscopic image of engraved lines, (d) Microscopic image of printed line (GC5 at 0.5 m/s)

The channels of the engraved lines have a width of about 47.5  $\mu\text{m}$  and a depth of about 18  $\mu\text{m}$  (see figure 5 (a), (b) and (c)). The printed line widths which could be reached with the printing form and a printing speed of 0.5 m/s are around 83  $\mu\text{m}$  which can be seen on the microscopic image in figure 5 (d).

The printed lines have higher widths than theoretically expected. This could be caused by the viscosity of the ink.

Also the intersections of the printed lines are analyzed. Figure 6 displays a comparison between the digital intersection and the printed one.

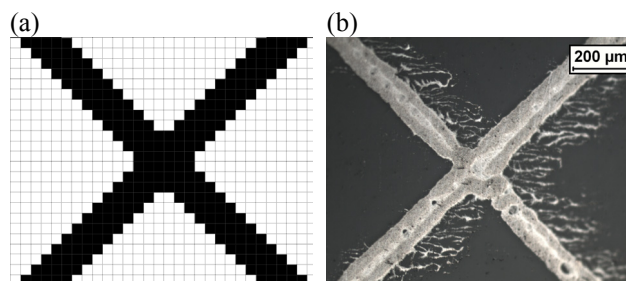


Figure 6: Comparison of cross-sections. (a) Cross-section of digital lines, (b) Cross-section of printed lines

It can be seen that the corners of the printed intersections are rounded.

Furthermore shadowing in print direction could be seen in the microscopic images. This effect occurs because of ink which is dragged outside of the cavities during the doctor blade movement. When the doctor blade moves further a fraction of this ink is deposited on the cylinder and transferred to the substrate. The shadowing effect depends on the printing speed and the viscosity of the used ink [7][8]. A comparison between the printing speed of 0.3 m/s and 0.5 m/s shows that the shadowing is reduced by higher printing speeds (Figure 7).

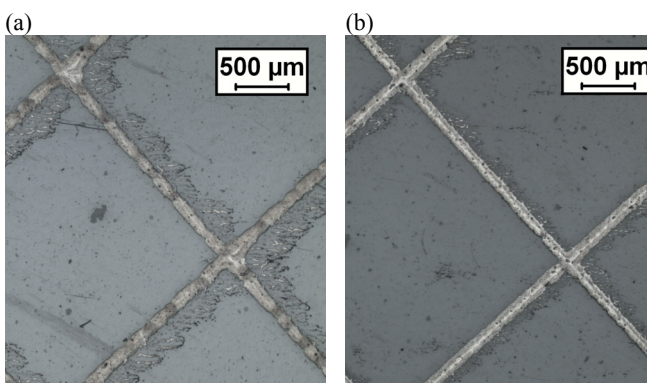


Figure 7: Comparison of shadowing at two different printing speeds. (a) Printing speed of 0.3 m/s, (b) Printing speed of 0.5 m/s

After printing, the grids are physically and electrically characterized. Figure 8 shows the average line heights and the standard deviation realized with three different printing speeds.

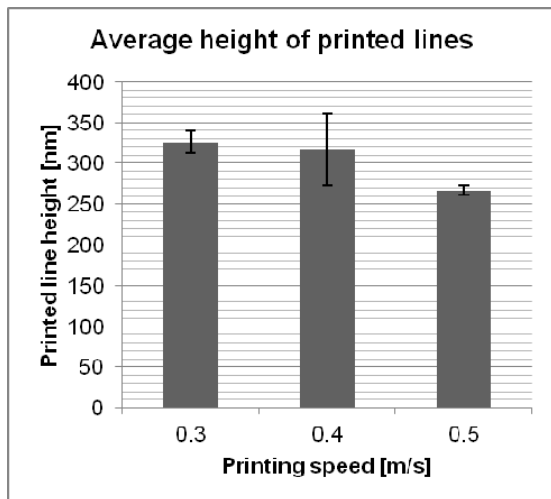


Figure 8: Average height of printed lines at printing speeds of 0.3, 0.4 and 0.5 m/s.

The diagram shows a decreasing line height with increasing printing speed.

After the physical characterization a measurement setup for the electrical analysis of the printed grids is determined. Previous surface resistance measurements were executed on single lines but a method for determining the sheet resistance of the whole grids is necessary.

Therefore the four point measurement by Van-der-Pauw is introduced on a grid area of 20x20 mm. For the measurement very thin needles are used. The average sheet resistances with standard deviations for the five grid constants printed with three different printing speeds are demonstrated in the diagram seen in figure 9.

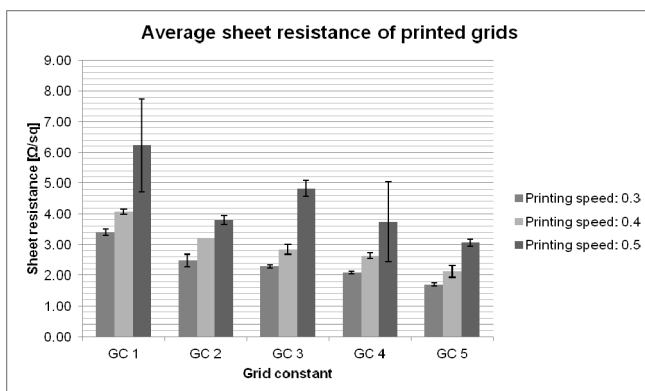


Figure 9: Average sheet resistance of printed grids printed with three different printing speeds

The diagram demonstrates that the sheet resistances at all printing speeds decrease with smaller grid constant. It can also be seen that the sheet resistance increases by raising the printing speed. This is a result of smaller layer thicknesses in case of higher printing speeds (see figure 8).

### Back injected components

The printed grids on PET foils are fixed on the wall of the cavity of the injection molding tool and back injected with thermoplastic polyurethane. The melt temperature is 260°C at an injection pressure of 740 bar and the mold temperature is kept constant at 40°C. Within the solidification a pre-pressure of 300 bar is set during a cooling time of 30 s.

Because of the stiff foil and high shrinkage of the polymer during injection molding, process related inter-laminar residual stresses between foil and polymer occur. This results in warping of the substrate in case of one-sided back injection of foil as it can be seen in figure 10.

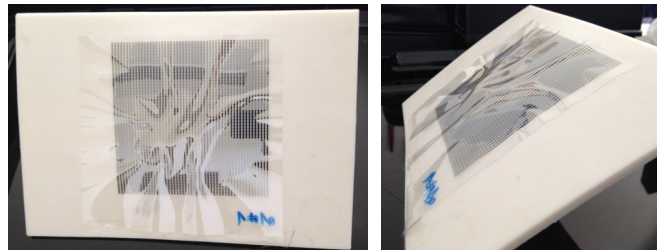


Figure 10: Back injected printed foil. (a) Front side of substrate with partial flaking of foil, (b) Little warping effect of back injection molded substrate

### Conclusion

Gravure printed silver patterns are printed with different grid constants and IR sintered. They should be applicable in back-injection-molded functional lightweight structures.

The height of the printed structures decreases with increasing printing speed but the sheet resistance of the grids rises by smaller layer thicknesses.

Optimizations in the printing form manufacturing have to be done regarding the intersections of the lines. Regarding the expected line widths the viscosity of used silver inks and the printing parameters have to be taken into account to realize lines as thin as possible.

The back injection and integration of the printed foils into light weight structures would be possible but this process step also needs further investigations regarding a smooth surface with a stable contact over the whole foil area. The optically and electrically characterization of the finished composites has to be done subsequent to the injection molding process.

### Acknowledgments

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## Author Biography

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