



# Fully roll-to-roll processed organic top gate transistors using a printable etchant for bottom electrode patterning



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## ABSTRACT

We demonstrate a high throughput roll-to-roll process for preparing organic top gate transistors with metallic electrodes on a flexible substrate. The source and drain electrodes are prepared by patterning a roll-to-roll evaporated silver layer with a printable etchant. The etching step is done by a screen printing process with a novel triblock copolymer based etching ink, which enables etching in just a few seconds at low temperatures. The method allows 100 μm resolution in the roll process and can produce thin (35 nm), smooth and nonporous electrodes with high conductivity. In addition, the process is independent on the printing direction and area, providing freedom of design. The polymeric semiconductor and insulator layers are deposited with roll-to-roll gravure printing, which allows orientation – and thus performance – of the semiconducting polymer to be controlled by the cup size and printing direction. The gate is printed with rotary screen, using a solvent-free silver paste as the ink. The properties of the prepared organic transistors are typical for fully printed devices: mobility of 0.017–0.026 cm<sup>2</sup>/(V s) and ON/OFF-ratio of 10<sup>2</sup>–10<sup>3</sup> were reached and the leakage current was very low (<0.1 nA).

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## 1. Introduction

The printability of organic semiconductor and insulator materials and their use in transistors has been demonstrated successfully in many cases [1–4] – arousing interest for their use in low-cost flexible electronics. One of the main targets in the organic thin film transistor (OTFT) research is to increase the stability and performance of organic transistors [5–7]. However, to fully exploit the potential for low manufacturing costs, the complete process has to be stable and transferred to a high throughput roll-to-roll (R2R) environment. One of the bottle necks in

fully R2R printed transistors is the manufacturing step for high quality bottom electrodes. Recently, some R2R compatible electrode patterning techniques have been demonstrated e.g. for printed transistors [8] and solar cells [9]. However, often the electrodes have been prepared by photolithography or other non-printing methods due to difficulties to prepare thin and smooth electrodes by printing, especially with high resolution and conductivity. The smoothness of the bottom electrode is essential for reliable transistor operation since the subsequent printed layers are very thin. The printed semiconductor layer can be as thin as 100 nm, which requires low thickness for the bottom electrodes in order to avoid problems e.g. with contact resistance [10]. Also the risk for short circuits increases with the electrode thickness since the thickness of the

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insulating layer cannot be increased infinitely without sacrificing the transistor performance, due to loss of field induced charges as the electric field decreases over the dielectric.

A few methods have been presented for R2R prepared electrodes. The simplest one is to print the electrodes with a metallic nanoparticle or nanowire ink. Approximately 20  $\mu\text{m}$  wide and 200 nm thick lines can be printed with standard printing methods (e.g. flexography and inkjet printing) if the process parameters are optimised carefully. However, the roughness, thickness and especially the spikes of directly printed metal lines often cause problems leading to short circuits. To overcome these issues at least one of the subsequent layers has to be carefully designed to have tolerance for peaks and spikes. Also other materials than metals, e.g. PEDOT:PSS [11] and graphene [12], can be used as directly printed electrodes for OTFTs. PEDOT:PSS electrodes have been shown to work e.g. in R2R gravure printed transistors [13] but conducting polymers cannot provide as good conductivity and stability against moisture as metals. On the other hand, R2R printed graphene is still suitable only for the top electrode since the current graphene inks are not able to provide sufficient conductivity as  $\sim 100$  nm thick layers, which are required for the bottom electrodes in OTFTs.

Other promising R2R-compatible electrode production methods include femtoliter gravure printing [14], flexo printing and lift-off [15], direct writing/filamentary printing [16],  $\mu$ -contact printing and etching [17], continuous phase-shift lithography [18], and screen printed, embedded electrodes [19]. In addition, nanoimprinted channels filled with metal nanoparticle ink [20] might provide a solution if the process can be transferred to a R2R environment. Also several electrode patterning methods based on surface energy control have been developed (see e.g. [21]) but the methods are typically slow and the transfer to a R2R compatible process is challenging [22].

In order to fulfil the requirements for high throughput, low cost electronics, a simple method to produce thin and smooth electrode patterns is needed. We introduce here a fast R2R process for patterning R2R evaporated metal films, which can be used as electrodes for many organic electronics applications, e.g. organic solar cells and transistors. The process is based on our ITO etching protocol [23] but instead of a commercial etching paste, we utilise a novel etching gel as the ink, which can be used as a screen printing paste with improved printability, higher etching speed and lower temperatures when compared to traditional printing pastes to pattern thin silver films. The ink consists of an etchant and a triblock copolymer, which forms a gel at elevated temperatures and/or at high concentration, both dissolved in water. The gelation allows accurate patterns to be printed since the ink solidifies when heated just after the screen printing step – thus hindering spreading of the ink.

The benefits of the R2R etching method include:

- ability to prepare thin electrodes  $\rightarrow$  minimise the risk for short circuits,
- freedom of design  $\rightarrow$  any arbitrary shape can be etched,

- direct etching  $\rightarrow$  only one printing step, no photolithography or etching resist is needed,
- no organic solvents  $\rightarrow$  water is used as a solvent and as a washing solution,
- rotary screen etching, washing and drying in just one run  $\rightarrow$  fast and continuous process (7 m/min printing speed, etching completed in a few seconds at low temperatures).

Finally, we present a complete R2R process for fully printed top gate organic thin film transistors, where each layer has been deposited in a continuous roll process. The transistors consist of the etched source and drain Ag electrodes on a polyethylene terephthalate (PET) roll, gravure printed polymeric semiconductor and insulator layers and a rotary screen printed Ag gate electrode on top.

## 2. Experimental

### 2.1. Materials

The etching ink consists of a triblock copolymer poly(ethylene oxide)–poly(propylene oxide)–poly(ethylene oxide) (PEO–PPO–PEO), i.e. Pluronic from BASF, and ferric nitrate ( $\text{Fe}(\text{NO}_3)_3$ ) from Aldrich as an etchant, both dissolved in water. In addition, an antifoam agent, Foam-Star<sup>®</sup> SI 2213 from BASF, was added to the ink in order to prevent foaming in the rotary screen process. The polymeric semiconductor ink in organic solvents (product GRAPE114) was received from BASF. Poly(methyl methacrylate) (PMMA) from Aldrich was used as the insulating polymer and was printed from a 7.5 wt% solution in organic solvents. The gate was printed using a solvent-free Ag paste (product IPC-114) from Inkron. A 125  $\mu\text{m}$  thick and 30 cm wide heat stabilized polyethylene terephthalate roll with an adhesion promoting coating from DuPont Teijin Films was used as a substrate.

### 2.2. Process

A continuous 35 nm thick silver layer was evaporated with a R2R evaporator on the PET substrate. The printability of the etching ink was first tested with a screen printing machine (EKRA E2) in a sheet-to-sheet (S2S) process and the process was then transferred to a rotary screen printing line. The rotary screen process was then optimised in order to find the best printing speed, drying temperature and time, concentration of the ink components and correct process for the washing step. Printability of the polymeric inks, i.e. semiconductor and insulator, was also first optimised in a S2S gravure printing process with Labratester from Norbert Schläfli Maschinen. After finding the best parameters in a sheet process, the process was transferred to a R2R gravure printing line. The Ag gate electrodes were first deposited by the EKRA E2 S2S screen printer in order to find out the optimal mesh and drying parameters and to check the compatibility of the Ag paste with the underlying layers. Finally, rotary screen printing was used to print the gate electrodes. Fig. 1 shows a schematic of the complete process, including the drying and washing steps. In

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