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Characteristics of flexographic printed indium–zinc-oxide thin films as an active semiconductor layer in thin film field-effect transistors

Stefan Dilfer[a,*], Rudolf C. Hoffmann[b], Edgar Dörsam[a]

[a] Institute of Printing Science and Technology, Technische Universität Darmstadt, Germany
[b] Department of Chemistry, Eduard-Zintl-Institute, Inorganic Chemistry, Technische Universität Darmstadt, Germany

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ABSTRACT

Characteristics of oxide semiconductor thin film transistors prepared by flexographic printing technique have been studied. The device was a field-effect transistor substrate (15 mm × 15 mm, n-doped silicon, 90 nm SiO_2 layer) with pre-structured gold electrodes and a printed active layer. The active layer was printed with a indium–zinc-oxide precursor solution and then annealed at 450 °C for 4 min on a hotplate. Influences of typographical parameters, i.e. printing pressure, anilox roller pressure, ink supply rate, printing velocity and printing plate (cliché) properties were studied. Reference active layers were produced by spin coating. The printed IZO ceramic layer with a dry film thickness between 3 and 8 nm, deposited onto the substrate for field-effect transistors provided a good performance with charge carrier mobilities (μ) up to 2.4 $cm^2 V^{-1} s^{-1}$, on/off current ratios ($I_{on/off}$ ratio) up to 5.2×10^7 and mean threshold voltages (V_{th}) of +4 V. The characterization of the printed and annealed IZO layer by AFM revealed the amorphous nature of the printed active layer films with a root-mean square roughness of 0.8 nm.

1. Introduction

Printing technologies as production methods for electronic devices are well-known in the silicon solar cell industry [1,2] and during the last decade even in the electronic industry. The possibility to produce polymer based thin film transistors, solar cells and displays as well as batteries by printing technologies have been previously examined [3–6]. From the industry side there is great interest to provide printed electronics as a low-cost mass production for the global economic market, as these new products have great economic potential [7–9]. Generally, any conventional or digital printing methods can be used to manufacture electronic components by printing technology. The selection of the printing method depends essentially on the specific properties of the function material to print [10,11]. Still it is possible to manufacture electronic components onto rigid and flexible substrates, such as glass, paper and plastic. Nowadays inorganic printable materials are one of the most intensively investigated materials in research topics today, especially in the area of printable production of thin film field-effect transistors (FETs). Particular amorphous oxide semiconductors find great interest in the chemistry, physics and material science communities, due to its high charge carrier mobility and $I_{on/off}$ ratio compared to organic semiconductors [12–22]. In this article, we have studied characteristics of oxide semiconductor field-effect transistors with active semiconducting indium–zinc-oxide (IZO) layer [21,22]. The precursor of indium–zinc-oxide, has been applied by flexographic printing technique onto substrates for FET devices with subsequent rapid drying at 450 °C.

2. Experimental

2.1. Synthesis of IZO precursor

The indium–zinc-oxide precursor consists of the composition indium–zinc 60/40 and was prepared according to Hoffmann et al., by a solution process using a combination of the two molecular precursors diaqua-bis[2-(methoxyimino)-propanoato] zinc(II) and tris[2-(methoxyimino)-propanoato]indium(III) [21].

2.2. Substrate

Substrates (1.5 cm × 1.5 cm) for FET devices (Fraunhofer IWS Dresden) consisted of n-doped silicon with a 90 nm layer of SiO_2, on which gold electrodes were attached with an intermediate adhesive layer of indium–tin oxide. The electrodes possessed an inter-digital structure with a channel width of 10 mm and a channel

* Corresponding author. Tel.: +49 263272459.
E-mail address: sdilfer@gmx.de (S. Dilfer).

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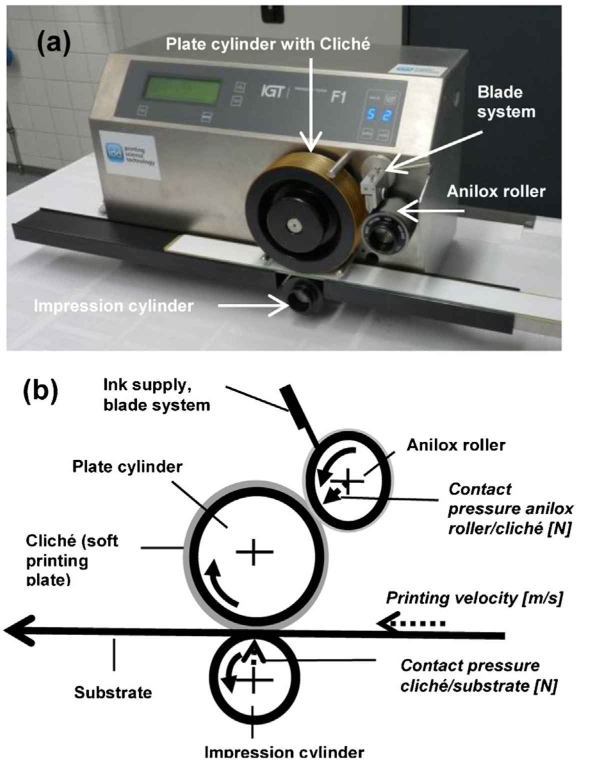

Fig. 1. (a) Printing experiment setup. (b) Scheme of the flexographic printing process.

length of 10 μm [18] (Fig. 5c and d). FET substrates were cleaned by consecutive ultrasonication in acetone, deionised water and iso-propanol for 15 min each to obtain a clean and residue free surface. The substrates were dried in a flow box for at least 1 h before being plasma treated for 1 min in an air plasma induced by a PFG300RF generator at 70 W [21,22].

2.3. IZO films preparation

Films were prepared by a printability tester F1 for flexographic printing from IGT Testing Systems (Fig. 1a). After printing the indium–zinc-oxide precursor onto the substrate for FET devices they were annealed with hotplate at 450 °C for 4 min which leads to the conversion of the precursor into an IZO ceramic. In this study, both the thicknesses of the IZO ceramics and the electrical properties of the fabricated FETs as function of the typographically flexographic printing parameters were investigated. As output parameters a contact pressure anilox roller/cliché of 10 N and a contact pressure cliché/substrate of 10 N were selected. The printing velocity was 0.8 m s^{-1}. A cliché of type nyloflex ACE114 of Flint Group with a shore hardness of 62 (A scale, DIN 53505) and a surface image of 60 L cm^{-1} (Lines per centimeter) and a screen frequency with 90% halftone area was used. An anilox roller type 402–209 with a cell volume of 20 mL m^{-2} and a screen frequency of 40 L cm^{-1} from IGT Testing Systems was used in the printability tester F1 as reference anilox roller. The typographic parameters were studied with three different values, respectively. Each experimental series was repeated five times with identical typographical parameters. The details of the investigated typographical parameters are as follows:

- 10, 50, 100 N contact pressure anilox roller/cliché,
- 10, 50, 100 N contact pressure cliché/substrate,
- 0.3, 0.8, 1.3 m s^{-1} printing velocity,
- anilox roller type 402–209 (20 mL m^{-2}, 40 L cm^{-1}), 402–213 (11.5 mL m^{-2}, 80 L cm^{-1}), 402–204 (8.5 mL m^{-2}, 100 L cm^{-1}) from IGT Testing Systems,
- and clichés nyloflex FE114 shore hardness 48 (A scale, DIN 53505, solid image), nyloflex FAH114 shore hardness 60 (A scale, DIN 53505, solid image), nyloflex ACE114 shore hardness 62 (A scale, DIN 53505, 90% halftone, 60 L cm^{-1} screen frequency image) from Flint Group.

As a reference FETs were produced by spin coating (30 s at 3500 rpm; no pre-spinning) of the appropriate precursor solution onto the substrates for FET devices.

2.4. IZO characterization

For measuring the IZO layer thickness a drop of 10% solution sulphuric acid removed the IZO ceramic from a substrate-corner. The acid was subsequently removed with a thin deionised water stream on the substrate and dried with argon. By means of an argon ion gun (operated at 250 eV and 500 nA) the samples were covered with an Au Layer by sputtering with a thickness of about 20 nm [11]. The IZO layer thicknesses were measured with a white light interferometer, NewView 6300 from Zygo. The film density and surface roughness of the IZO ceramic were investigated by atomic force microscopy (AFM): CP-II (Brucker-Veeco), with a scan rate of 0.5 Hz, using silicon cantilevers [21]. FET characteristics were determined with a Agilent 4155A Semiconductor Analyzer in a glove box, in daylight under argon atmosphere. Charge carrier mobility μ, on/off current ratio $I_{on/off}$ *ratio* and threshold voltage V_{th} were derived from a linear fitting of the square root of the voltage of the source drain current ($I_{DS}^{0.5}$) as a function of gate source voltage V_{GS} [18,21].

3. Results and discussion

Various printing technologies such as ink jet printing, screen printing, gravure printing, flexographic printing and offset printing have been utilized for printed electronics [11,23,24]. Compared to other contact printing technologies, flexography requires only a slight contact pressure to enable reliable ink transfer from the cliché to the substrate. This contact pressure must be as even as possible on all printing locations along the contact area and during the pass of the entire print length to guarantee a good printing quality. Still it is possible to print on very thin, flexible and solid substrates, virtually all papers, thick cardboard, rough-surfaced packaging materials and fabrics, which is what differentiates flexographic printing to other contact printing techniques [25]. Flexographic printing is a relief printing technology with raised printing elements. The clichés, flexible printing plates, composed of rubber or plastic are attached to the plate cylinder. Viscosity of inks used for flexographic printing is typically in a range from 50 up to 500 mPas and thereby higher than that for ink jet printing but lower than that for screen and offset printing [10]. The printing plate is inked with a screen roller, often called an anilox roller, which is evenly patterned with cells. A blade system assures the equal filling of the cells on the anilox roll and therefore stabilizes the printing process. With the inked cliché, the ink is transferred onto the substrate with the aid of an impression cylinder (Fig. 1a and b). By using a low pressure between the plate cylinder and impression cylinder the ink is transferred to the substrate [10].

3.1. Film thickness

The white light interferometer images shows an IZO edge were the film thickness was measured (Fig. 2a) and a mean IZO layer film