Organic Electronics 39 (2016) 168–176

Contents lists available at ScienceDirect

Organic Electronics

journal homepage: www.elsevier.com/locate/orgel

All-inkjet-printed dissolved oxygen sensors on flexible plastic substrates

Ana Moya [a, d], Enrico Sowade [b], Francisco J. del Campo [a], Kalyan Y. Mitra [b], Eloi Ramon [a], Rosa Villa [a, d], Reinhard R. Baumann [b, c], Gemma Gabriel [a, d, *]

[a] Instituto de Microelectrónica de Barcelona, IMB-CNM (CSIC), Esfera UAB, Campus Universitat Autònoma de Barcelona, 08193, Bellaterra, Barcelona, Spain
[b] Technische Universität Chemnitz, Digital Printing and Imaging Technology, 09126, Chemnitz, Germany
[c] Fraunhofer Institute for Electronic Nano Systems (ENAS), Printed Functionalities, 09126, Chemnitz, Germany
[d] Research Networking Center in Bioengineering, Biomaterials and Nanomedicine (CIBER-BBN), 08193, Barcelona, Spain

ARTICLE INFO

Article history:
Received 28 June 2016
Received in revised form
29 September 2016
Accepted 2 October 2016

Keywords:
Inkjet printing
Oxygen sensors
Microelectrodes
Plastic substrates
Low-cost

ABSTRACT

Inkjet printing is a promising alternative manufacturing method to conventional standard microfabrication techniques for the development of flexible and low-cost devices. Although the use of inkjet printing for the deposition of selected materials for the development of sensor devices has been reported many times in literature, it is still a challenge and a potential route towards commercialization to completely manufacture sensor devices with inkjet technology. In this work is demonstrated the fabrication of a functional low-cost dissolved oxygen (DO) amperometric sensor with feature sizes in the micrometer range using inkjet printing. All the required technological steps for the fabrication of a complete electrochemical three electrodes system are discussed in detail. The working and counter electrodes have been printed using a gold nanoparticle ink, whereas a silver nanoparticle ink was used to print a pseudo-reference electrode. Both inks are commercially available and can be sintered at low temperatures, starting already at 120 °C, which allows the use of plastic substrates. In addition, a printable SU8 ink formulation cured by UV is applied as passivation layer in the sensor device. Finally, as the performance of analytical methods strongly depends on the working electrode material, is demonstrated the electrochemical feasibility of this printed DO sensor, which shows a linear response in the range between 0 and 8 mg L^{-1} of DO, and affords a detection limit of 0.11 mg L^{-1}, and a sensitivity of 0.03 μA L mg^{-1}. The use of flexible plastic substrates and biocompatible inks, and the rapid prototyping and low-cost of the fabricated sensors, makes that the proposed manufacturing approach opens new opportunities in the field of biological and medical sensor applications.

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1. Introduction

Inkjet printing is attracting increasing interest in the area of micro-sensors technology, e.g. for the determination of biological or chemical parameters with a high spatial resolution [1,2,3]. The inkjet printing method is considered a promising alternative to traditional sensor manufacturing techniques, as it is an additive and non-contact approach that allows the mask-less deposition of different functional materials on rigid and flexible substrates. Even though screen-printing still remains the most used printing technique employed for the production of low-cost and disposable sensors [4,5], the emerging inkjet printing technology has many advantages over it, such as easily changeable layouts without the need of physical stencils or screens, the reduction of the generated waste material and the non-contact deposition. The latter feature helps prevent substrate contamination, and it allows the technique to be used on delicate substrates that may be damaged by the force applied in the screen-printing process. Moreover, the direct writing approach without any need for screens drastically reduces the overall fabrication time and costs of an entire device, and also facilities iterative design changes during development. Therefore, inkjet printing can be ideal in prototyping as well as in applications requiring heterogeneous integration of different components and materials [6]. Several attempts have been made until now using inkjet printing for the fabrication of printed chemical sensors.

* Corresponding author. Instituto de Microelectrónica de Barcelona, IMB-CNM (CSIC), Esfera UAB, Campus Universitat Autònoma de Barcelona, 08193, Bellaterra, Barcelona, Spain.
E-mail address: gemma.gabriel@imb-cnm.csic.es (G. Gabriel).

http://dx.doi.org/10.1016/j.orgel.2016.10.002
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However, the number of reports about biosensors in flexible devices is still limited. Some of the first inkjet-printed biosensors were reported back in 1992 by Newman et al. [7] demonstrating the usefulness of the inkjet technology by printing a glucose sensor. More recently, other works have been presented, such as those by Jensen et al. [2], providing the viability of detecting a cancer biomarker in serum by printing an array of gold electrodes functionalized with 3-mercaptopropionic acid, or by Khan et al. [8] reporting a flexible gold electrode array to be used as bioelectronic interface. Furthermore, many other publications can be found on electrochemical biosensors [9,10], pH sensors [11,12], ion sensors [13], gas and vapor sensors [1,14–16] and photosensors [17] electromyography biosensors [18]. However, most of the printed sensors reported in the literature have only some of their elements printed while others are still manufactured by traditional lithography-based or related methods.

We recently demonstrated the fabrication of an electrochemical micro-sensor for the measurement of dissolved oxygen (DO) using standard microfabrication techniques [19]. Microfabrication yields highly reproducible and reliable microelectrodes with a sensing response comparable to commercially available dissolved oxygen sensors [20]. However, these standard microfabrication techniques require multiple complex and time consuming processing steps including the application of photoresist, UV-light exposure through chromium masks, etching and deposition of adhesion and electrode materials. The nature and high number of process steps increases the cost per micro-sensor, thus limiting their application as disposable devices.

The aim of this work is to demonstrate the potential of inkjet printing technology as an alternative to standard microfabrication techniques in the area of micro-sensors fabricating on a flexible plastic substrate. Polymeric substrates such as PEN are promising candidates for disposable low-cost sensors. They are flexible, inexpensive, and available in large quantities, facilitating roll-to-roll processing [21]. However, usually they are sensitive towards high temperatures due to their comparable low glass transition temperature. Therefore, all required manufacturing processes have to be performed at low temperatures, preferable much below 200 °C. Although gold is one of the most important electrical conductive materials for a wide range of applications, the processing of the inkjet printing inks is still a challenge due to the high sintering temperatures reported until now. Different strategies have been developed for the drying and sintering process to limit the temperature effect on polymer substrates such as the usage of intense pulsed light [22], laser [23] or infrared technology [24,25]. Since there are rarely Au ink formulations on the market that can be sintered at low temperatures compatible with low Tg polymer substrates, several authors overcome these problems synthesizing their low-curing inks [2,26,27]. However the stability and reproducibility of these custom-made inks is still limited [28,29]. Furthermore, the synthesis of these inks is difficult, and not always in the field of expertise of research groups that are focused on the final application. Nowadays, ink materials are the most important drawback that limits inkjet printing technology to be widely used. This especially affects to novel materials such as gold and platinum. However, silver nanoparticle inks are state-of-the-art. There is currently a large variety of silver nanoparticle inks for different applications commercially available [30].

We aim to develop an all-inkjet-printed sensor using stable and commercially available inks that can be processed and sintered at low temperature (120 °C). These inks allow the usage of flexible Polyethylene Naphthalate (PEN) substrate and are commercial and reliable in terms of stability. The inks are used to develop a fully inkjet-printed oxygen amperometric sensor using different metal nanoparticle inks and a dielectric ink formulation (gold for the counter (CE) and working electrode (WE); silver for the pseudoreference electrode (pRE), and the dielectric SU8 as passivation). Our proposed manufacturing strategy is very attractive due to its simplicity and rapidity, and the good performance of the microelectrodes through their lifetime. Taking advantage of the low-cost provided by inkjet printing, these electrochemical platforms presented here potentially enable novel devices in a broad range of applications such as analytical sensors, biosensing and medical applications.

2. Material and methods

2.1. Materials and chemicals

For the development of the DO sensor we used three commercially available inks. A low-curing gold nanoparticle ink (Au-LT-20 from Fraunhofer IKTS, Germany) was employed for the WE and CE development. A silver nanoparticle ink (DGP-40LT-15C from ANP, Korea) was used for the development of the pRE electrode. The passivation of the electrodes was done using the SU8 ink (2002 from MicroChem, USA). The inks show drop-on-demand (DoD) inkjet compatible specifications presented in Table S1 (Supplementary Information). Teonex Polyethylene Naphthalate PEN films (Q65HA DuPont Teijin Films) with a thickness of 125 μm and a surface pre-treatment for improved adhesion was selected as substrate.

Ethanol (LC/MS grade), sodium nitrate (KNO_3), potassium hexacyanoferrate(III) ($K_3[Fe(CN)_6]$) and Potassium hexacyanoferrate(II) trihydrate ($K_4[Fe(CN)_6]$) (all from Sigma Aldrich, Spain) were used for surface cleaning, activation and characterization of the printed sensor. Hydrochloric acid (0.1 M) was electrochemically applied for the chlorination of the printed silver layers, and potassium chloride (KCl) for testing the open circuit potential of the pRE (both from Sigma Aldrich, Spain).

2.2. Instrumentation

A piezoelectric Dimatix Material Printer (DMP 2831 from FUJIFILM-Dimatix, Inc., USA) was employed for the inkjet printing of the three inks on the PEN substrate. The printer was equipped with user fillable 10 pL nominal drop volume printheads having 16 nozzles each with a diameter of 21.5 μm. Printing patterns were made using Electronic Design Automation (EDA) layout software and imported with the Dimatix Bitmap editor software. The substrate vacuum plate is temperature controllable. The printing processes were carried out in a standard laboratory environment in ambient condition, without non-particulate filtered enclosure systems and without control of temperature or humidity to determine the extent to which the sensor devices could be manufactured on an industrial-scale printing system.

Scanning Electron Microscopy (SEM, Auriga-40 from Carl Zeiss) was used to study the morphology of each printed layer. An Atomic Force Microscope (AFM, Nanoscope IV controller and Dimension 3100 head from Veeco) was employed to analyze the topography of the printed layers and a profilometer Dektak 150 (Veeco) was used for the thickness measurement of the layers. The sheet resistance of each conductive layer was determined with a Hewlett Packard HP4155 Semiconductor Parameter Analyzer connected to a manual probe station PM5 (Süss Microtec Gmbh). The contact angle measurement system MobileDrop GH11 (Krüss Gmbh) was used to investigate the surface energy of the substrate.

The electrochemical characterization and calibration of the sensors was performed with an 8-channel potentiostat 1030A Electrochemical Analyzer (CH Instruments, USA). Control experiments were performed using a commercial Ag/AgCl (3 M KCl)

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