Sensors and Actuators B 268 (2018) 123–128

Contents lists available at ScienceDirect

Sensors and Actuators B: Chemical

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

Transparent In-Ga-Zn-O field effect glucose sensors fabricated directly on highly curved substrates

Xiaosong Du, Gregory S. Herman*

Oregon State University, School of Chemical, Biological, and Environmental Engineering, 116 Johnson Hall, Corvallis, OR, 97331, USA

ARTICLE INFO

Article history:
Received 21 December 2017
Received in revised form 15 April 2018
Accepted 18 April 2018
Available online 22 April 2018

Keywords:
Transparent biosensor
a-IGZO glucose sensor
a-IGZO thin-film transistor
Microcontact printing

ABSTRACT

Fully transparent electronics are of increasing interest for biological applications, where the combination of sensing and imaging can potentially improve patient healthcare diagnostics. Herein, we report high-performance, fully-transparent amorphous In-Ga-Zn-O field-effect transistor (a-IGZO FET) based biosensors fabricated directly on highly-curved glass substrates. The a-IGZO channel and indium tin oxide electrodes were patterned directly on glass tubes (1.0 mm radius) by microcontact printing self-assembled monolayers and etching the metal oxide films. This approach led to a-IGZO FETs with excellent electronic performance, with high on/off drain current ~1.3×10^6, high average electron mobility > 7.4 cm^2/Vs, low on/off hysteresis ~0.6, V and low gate leakage current ~10^{-10} A. The back-channel of the a-IGZO FETs were functionalized with glucose oxidase to make fully transparent biosensors with very high sensitivity to glucose. We have determined that the glucose limit of detection is 170 μM. These results provide insight into new methods for fabricating a-IGZO FETs and a-IGZO FET biosensors on non-planar substrates, and may open a range of new applications, including transparent sensing catheters, flexible active transparent electrode sensing arrays, and integration of FET based biosensors on optical fibers.

1. Introduction

Metal oxide and carbon nanomaterials have the potential to make high-performance, transparent, active electronic devices [1–5]. Much of the initial interest in transparent thin film transistors was towards their application in relatively simple circuits and transparent displays [6]. More recently, there has been considerable interest in the combination of transparency with sensing for a range of novel biosensing applications, including simultaneous electrophysiological recordings and neural imaging [7,8], integrated bioelectronics on an endoscope [9], smart contact lenses [10–13], and pressure sensors for medical applications and soft robotics [14]. The basis of these devices is the integration of transparent electrodes or transparent transistors on flexible transparent substrates, which can then be transferred to a different substrate [15,16]. Often the flexibility of the final sensor array is not critical, but being able to integrate these sensors onto a new form factor makes novel and unique applications possible.

The integration of active electronics on curved surfaces has been explored, where deposition and patterning is performed on a planar surface which is then transferred to a curved surface [17] or allowed to elastically deform into the desired shape [18]. However, it has been difficult to directly generate patterns on curved substrates by traditional patterning technologies, including photolithography and e-beam lithography [19]. The use of soft lithography methods, such as microcontact printing (μCP), can pattern complex, three-dimensional topologies with submicron-scale features on curved surfaces [20,21]. This approach may be of great interest to large-scale manufacturing, especially for roll-to-roll or other high throughput processes [22–24]. μCP takes advantage of the ability of an elastomeric stamp to conform to non-planar substrates with minimal distortion of the printed pattern. Self-assembled monolayers (SAMs) transferred from a patterned stamp onto the surface results in a robust, dense protective layer that can be used as a chemical resist to prevent etching of the SAM covered material [25]. μCP has been shown to be a promising method to pattern transparent oxide films [26] and field effect transistors (FETs) [27,28].

There has been increasing interest in using transparent amorphous oxide semiconductors for flexible electronics [13,29,30]. Amorphous In-Ga-Zn-O (a-IGZO) is the most studied transparent amorphous oxide semiconductor, and is currently used as an active material in FETs for flat-panel displays [3,31–33]. A key benefit of a-IGZO for flexible electronics, is that low processing temperatures on flexible, polymeric substrates is possible, while retaining relatively high electron mobilities, low operating voltages, and very low off currents [10,34]. Recently, a-IGZO FETs have been widely

* Corresponding author.
E-mail address: greg.herman@oregonstate.edu (G.S. Herman).

https://doi.org/10.1016/j.snb.2018.04.087
0925-4005/

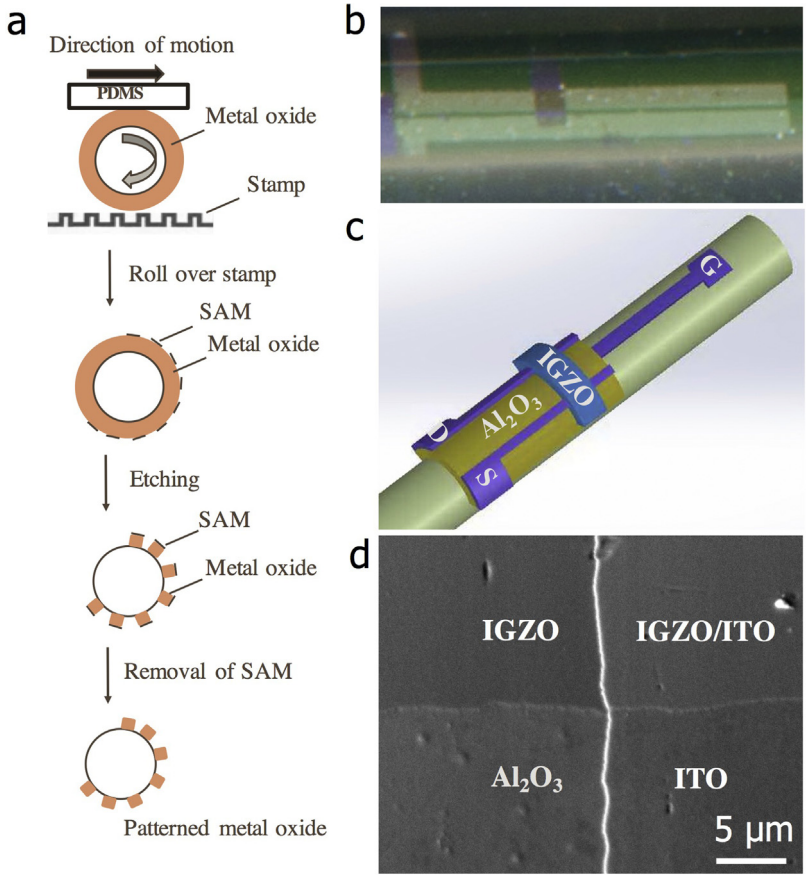

Fig. 1. (a) Schematic illustration of μCP on a curved substrate. (b) Optical image and (c) schematic illustration of a-IGZO FET structure on a curved substrate (W/L ratio of 100 μm/20 μm). (d) SEM image of the step edge between ITO and Al_2O_3, and between a-IGZO/ITO and a-IGZO.

studied for use as sensors, including gas detection [35], pH [36,37], temperature [38], light sensing [39], pressure sensing in contact lenses [10], and biochemical sensing [40–43]. Recently a-IGZO FET biosensors have been demonstrated as an efficient approach to detect glucose levels [44,45], and can potentially be used as a critical component of an artificial pancreas for diabetes care [46,47].

In this study, we demonstrate a novel method to fabricate fully-transparent, bottom-contact, bottom-gate a-IGZO FETs directly on highly curved substrates using soft lithography. Standard vacuum-based approaches were used to deposit high quality a-IGZO, In-Sn-O (ITO), and Al_2O_3 films. Using μCP we were able to pattern the a-IGZO and ITO films directly on 1.0 mm radius glass tubes to form the semiconductor channel and gate/source/drain electrodes, respectively. The electrical characterization of the fabricated a-IGZO FETs and biosensors are investigated and compared to prior studies on planar substrates.

2. Experimental section

2.1. Materials

Glucose was obtained from Alfa Aesar. HCl, NaCl, KCl, NaH_2PO_4, Na_2HPO_4 were acquired from Macron. Aminopropyltrimethoxysilane (ATPMS) was purchased from Sigma-Aldrich. Glutaraldehyde (GA) was acquired from Electron Microscopy Sciences. Glucose oxidase (GOx) was obtained from Amresco. Octadecylphosphonic acid (ODPA) was purchased from Specific Polymers. Tridecafluoro-1,1,2,2-tetrahydrooctyl-1-trichlorosilane (TFOCS) was purchased from UCT Specialties. The SU-8 photoresist was acquired from Microchem. IGZO and ITO targets were purchased from AJA International Inc. and Kurt J. Lesker Inc., respectively. Sylgard 184 polydimethylsiloxane (PDMS) was obtained from Dow Corning. Milli-Q water was used for sample preparation.

2.2. Fabrication of μCP stamp and well

An epoxy-based negative photoresist (SU-8) was spin-coated onto a clean silicon wafer. The photomask was aligned in close contact with the wafer and an ultra-violet light source (model 100UV30S1, Karlsus Inc.) with wavelength ~360 nm was used to expose the photoresist. Developer solution was used to remove unexposed regions of SU-8 from the substrate. The remaining SU-8 pattern on the silicon wafer (the master) had a thickness of approximately 50 μm as measured by profilometry (KLA-Tencor Alpha-Step 500). TFOCS was deposited as a monolayer on the master through siloxane bonding by placing the master in TFOCS vapor for 30 min. Liquid pre-polymer PDMS with curing agent (5:1 weight ratio) was cast onto the master followed by degassing for 30 min in vacuum. The PDMS was cured in an oven at 70 °C for 7 h. A fresh PDMS stamp with features opposite to the master was obtained by peeling the PDMS stamp from the master substrate. A similar procedure was used to fabricate the PDMS well, where SU8 was patterned as a $1 \times 2.5\ mm^2$ rectangle that was 100 μm thick, and the PDMS was cast onto the substrate. The PDMS well was then wrapped around the tube so that the well was located over the biosensor, and a syringe was used to deliver the various solutions to the device.

2.3. Device fabrication

μCP FETs were fabricated as follows, where the general procedure to pattern metal oxide films (both a-IGZO and ITO) on cylindrical surfaces using μCP is shown in Fig. 1a. A glass tube