# Flexible glucose sensor utilizing multilayer PDMS process

Jasbir N. Patel*, Bonnie L. Gray*, Bozena Kaminska* and Byron D. Gates**
*School of Engineering Science, Simon Fraser University, Burnaby, BC, CANADA.
**Department of Chemistry, Simon Fraser University, Burnaby, BC, CANADA.

***Abstract*— In this paper, a flexible glucose sensor with gold electrodes sandwiched between two polydimethyle siloxane (PDMS) layers is presented. PDMS is used as a flexible and bio-compatible sensor substrate material. Furthermore, PDMS is optically transparent and haemocompatible which is very important for implantable sensors.**

**The novel self registration fabrication process gives accurate alignment to all the layers of the glucose sensor and it is easy to fabricate without any manual interaction or alignment. Furthermore, crosslinked SU-8 (a negative photoresist) is used first time as a lift-off layer to pattern metal on the PDMS surface.**

**The bottom PDMS layer of the sensor is ~500 μm thick and the top PDMS layer is ~120 μm. Each sensor chip is 5.5 mm X 10.5 mm in size with 2 mm X 3 mm for the actual sensor area. Resistance of the successfully fabricated gold electrodes is measured between 3 to 6 mΩ. The drift of the sensors are measured using high purity water followed by the linearity test using hydrogen peroxide ($H_2O_2$).**

## I. Introduction

THE need for regular glucose monitoring was highlighted in a study by the Diabetes Control and Complications Trial Research Group (1993) in the USA [1]. Although early debate centered on whether there was a need for a long-term implantable glucose sensor for diabetics, recent long-term studies have conclusively demonstrated that if glucose levels can be tightly regulated within the normal physiological range, then diabetic complications can be controlled [2].

Many different types of glucose sensors have been developed and can be classified by different aspects of their design. Optical glucose measurement is widely used for portable hand held glucose meters [3]. However, this method gives only rough glucose concentrations instead of the exact measure of blood glucose. Glucose detection using the affinity principle [4] detects a change in the physical properties of a sensitive cantilever, usually a change in the resonant frequency due to mass loading.

The oldest electro-enzymatic detection principle, developed by Clark in the 1960's, is still of interest to many researchers because of its high selectivity to glucose. The stage of proof of concept for such a device has passed, and currently efforts are aimed at improving the performance of electro-enzymatic sensors.

First among the problems with an enzymatic glucose sensor is the unpredictability of the Ag/AgCl electrode fabrication and drift in current. Moreover, platinum is widely used as an active electrode but is quite an expensive material for large scale production of the glucose sensors.

Finally, flexibility and optical transparency is crucial for sensors that may be worn or implanted, such as within contact lenses in the eyes for detecting glucose in tears. However, a fully transparent sensor must be fabricated using optically transparent electrodes (e.g. ITO) and such a transparent sensor should be designed using a flexible material such as PDMS. The only design that is both optically transparent and mechanically flexible has been proposed by Mitsubayashi et. al.[5]. However, this design uses platinum or indium tin oxide (ITO) and silver/silver chloride electrodes.

In order to address some of the problems discussed above, a PDMS based flexible glucose sensor with optically transparent substrate is presented here. Gold as an electrode (both the active and the reference electrodes) material is presented for the first time. A simple fabrication process flow without any manual interaction is also presented. This hybrid polymer fabrication process uses SU-8 as a lift-off material for metallization on the PDMS surface.

## II. Design

The flexible electro-enzymatic sensor is fabricated using PDMS as a sensor material. PDMS is widely used for microfluidic and biomedical applications because of its ease of fabrication and biocompatibility [6]. PDMS is used as a sensor substrate which is optically transparent and flexible.

In addition, both the active and the reference electrodes of the flexible sensor are fabricated using only gold. Gold, similar to platinum, is also a biocompatible material [7] [8]. The motivation for further characterization of gold electrodes for the flexible sensors comes from the primary results obtained on a glass substrate [9].

The fabrication process can be used for batch fabrication of many sensors at once, which is crucial for commercialization of any glucose sensor. The sensor design avoids any manual assembly or alignment steps. The presented sensors are cost effective, simple to fabricate, and easy to release after complete fabrication.

In order to test and characterize different properties of the flexible electro-enzymatic glucose sensor, we designed a flexible sensor with sandwiched gold electrodes between two PDMS layers (Fig. 1). The bottom layer of the sensor is a 500μm thick rectangular PDMS layer. This bottom layer is little larger than the area occupied by the electrodes.

The top sensor layer is designed using a 100 to 200 μm

thick PDMS layer with contact pad openings and electrode openings (Fig. 1). The electrode opening is provided only in the sensing area. Two separate openings are designed so that any of the gold electrodes can be used as an active electrode by immobilizing the enzyme on them. And, as mentioned previously, both gold electrodes are sandwiched between the two PDMS layers.

All the sensor designs consist of three sections: a sensing area, a connecting conductor, and a contact pad. Each sensor die is 5.5 mm x 10.5 mm in size and is fabricated with gold electrodes sandwiched between two layers of PDMS (Fig. 1). In a single step metal patterning process, the active and the reference electrodes, the connecting conductors, and the contact pads were fabricated using only gold. It should also be noted that the sensor dimensions, including bond pad dimensions, were chosen for easy handling and testing; the batch fabrication process could be utilized for both larger and much smaller sensors.

The actual sensing area, in all the designs, is 2 mm x 3 mm (Fig. 1). However, the active and reference electrode areas are 2mm x 2mm in size.

Large contact pads are designed for each electrode to make wire connections easier. Each 2mm x 2mm contact pad is connected with a 6mm long gold conductor. The openings in the top PDMS layer for the contact pads and the sensing electrode (Fig. 1) are made for the physical connection with wires and for contact with the enzyme respectively. The gold conducting wire is covered with PDMS to avoid direct external contact (Fig. 1).

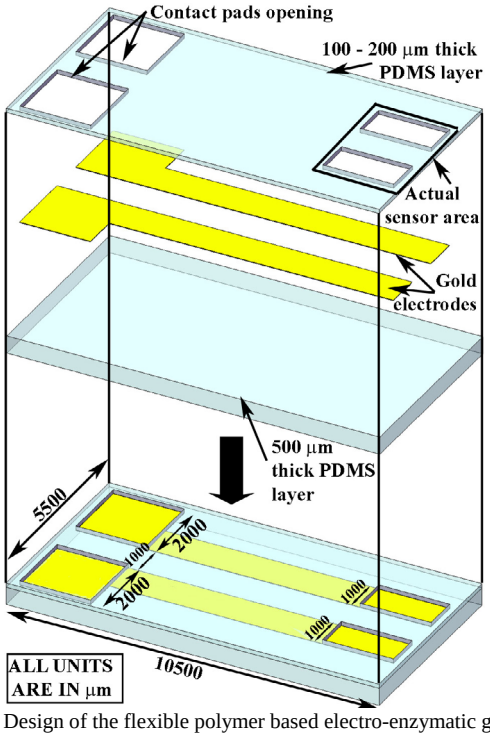


Fig. 1: Design of the flexible polymer based electro-enzymatic glucose sensor

## III. FABRICATION

The hybrid polymer fabrication process of the flexible electro-enzymatic glucose sensor is carried out using PDMS as the sensor substrate. Moreover, a thin SU-8 layer is used as a lift-off mask for metallization on the PDMS surface.

A hybrid polymer fabrication flow for the gold electrodes sandwiched between the two PDMS layers is presented in Fig. 2. Each step of the hybrid polymer fabrication flow is explained below.

- First of all, a cleaned glass substrate is spun with SU-8 2005 to make uniform coating on the glass substrate. This SU-8 layer behaves as a low adhesion material between the glass substrate and the bottom PDMS layer. The SU-8 2005 layer is soft baked, flood exposed and hard baked (Fig. 2(b)).
- To fabricate the bottom PDMS layer, Sylgard® 184 elastomer with 10% curing agent is mixed and is poured on the substrate. The mixture is cured at 85° C for 2 hours and 30 minutes (Fig. 2(c)).
- 100μm thick SU-8 2035 is spun on the PDMS layer and is soft baked by ramping the temperature. The SU-8 layer is exposed with the first mask to realize lift-off mask for the gold electrodes. The 365nm (i-line) UV source is used to expose the SU-8 layer. The SU-8 is baked again for a post exposure bake (to cross-link the polymer) by ramping the temperature. Finally, the unexposed SU-8 layer is developed using SU-8 developer until the unexposed SU-8 is completely dissolved (Fig. 2(d)).
- To make gold electrode patterns, 50nm chrome and 100nm gold layer are sputtered using the Corona sputtering system (Fig. 2(e)).
- Peel off the 100 μm thick SU-8 layer to leave the gold electrode patterns on the PDMS surface (Fig. 2(f)).
- Spin and bake thick photoresist (S1827). Sputter 50 nm chrome layer on the resist to avoid development during the SU-8 development process (Fig. 2(g)).
- Now, spin 100 - 200 μm thick SU-8 2100 layer. This layer is used to make the rings around the sensor active area and the contact pads in the top PDMS layer. Soft bake the SU-8 layer with ramping up the hot plate followed by the UV exposure of the layer. The development of the thick layer is done after post exposure bake on the hot plates (Fig. 2(h) and Fig. 2(i)).
- Etch the chrome layer using Transene chrome etchant followed by the flood expose of the photoresist layer. The photoresist layer can be removed either by MF 322 developer or by the SU-8 developer (Fig. 2(j)).
- The top PDMS layer is poured now and cured on hot plate. The top PDMS layer is very well attached to the bottom PDMS layer (Fig. 2(k)).
- The SU-8 rings, chrome and photoresist are finally removed by soaking the substrate in acetone. To save time, ultrasonics is used (Fig. 2(l)).
- Finally, the sandwiched PDMS sensors are peeled off from the process substrate and separated out using a knife (Fig. 2(m)).

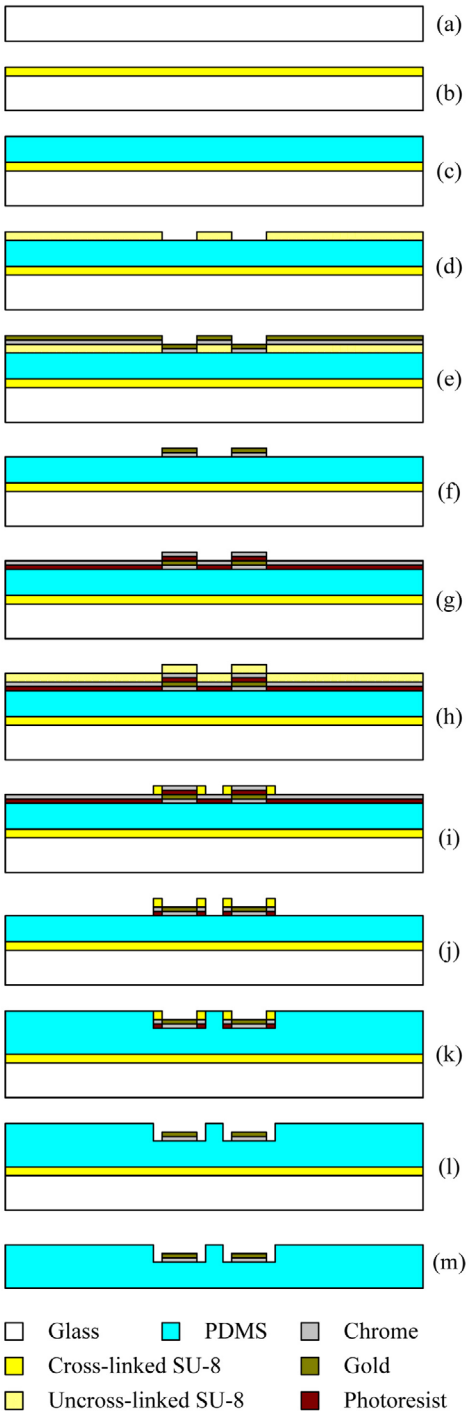


Fig. 2: Hybrid polymer fabrication process for the sandwiched gold electrodes between the two PDMS layers: (a) a cleaned glass substrate, (b) thin SU-8 2005 layer; (c) pour PDMS on the substrate and cure; (d) pattern SU-8 layer for lift off; (e) sputter chrome and gold; (f) peel off SU-8 layer to leave metal pattern on the PDMS surface; (g) spin photoresist and sputter Cr again as a releasing mask for SU-8 rings; (h) spin and pattern SU-8 2100 layer for SU-8 rings, (i) develop thick SU8 2100 layer, (j) etch chrome, flood expose photoresist and develop the photoresist, (k) pour PDMS to make top sensor substrate layer, (l) remove SU8 rings, chrome and photoresist by soaking in acetone using ultrasonics, and (m) peel off the sandwiched sensors.

Sensors are connected with wires using a conductive silver epoxy for testing. At this point, after optical verification, each sensor is ready for the electrical test.

## IV. EXPERIMENTATION

Characterization of our gold electrodes included analysis for consistency between each of the final sensors. We used a combination of optical microscopy and profilometry to monitor uniformity in dimensions and thickness of our flexible sensor array. Each sensor was tested for electrical conductivity before immobilizing the enzyme on them.

The response of the sensor without immobilizing the enzyme was tested using hydrogen peroxide ($H_2O_2$). The concentration of $H_2O_2$ is varied from 0 to 50 percent in the phosphate buffer to verify the correct functionality of the sensors. Potentiostat (Pine Instruments Inc.) is used to record the response of the sensors with increasing $H_2O_2$ concentration. The potentiostat is set to supply constant potential of 1 V.

## V. RESULTS AND DISCUSSION

The substrate with sensors was optically checked several times during the hybrid polymer fabrication process. After the electrodes were patterned on the first layer of PDMS using SU-8 lift-off, electrical conductivity of all the electrodes were satisfactorily checked. The results obtained from each test during the fabrication process and after the fabrication are discussed below.

### *A. Results related to fabrication process*

SU-8 is first time used as a lift-off material for metallization on the PDMS surface. The previous investigation of the process indicates that the required force to peel-off the SU-8 layer is very low [8]. The peeled SU-8 layer from the PDMS surface and the patterned electrodes on the PDMS surface are shown in Fig. 3. The electrical resistance of each electrode is between 3 and 6 mΩ.

After successful fabrication of each sensor, all the sensors are easily peeled from the SU-8 coated glass substrate. The electrical conductivity of each sensor is checked and the sensors with lost conductivity are discarded. A collection of dual layer sandwiched PDMS sensors are shown in Fig. 4. The conductivity measurement indicates uniform and defect-free gold electrodes and PDMS layers.

The flexibility of the PDMS based sensors are shown in

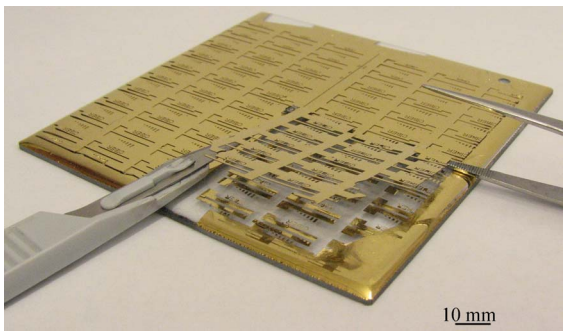


Fig. 3: Successfully peeled SU-8 layer and patterned gold electrodes on the PDMS surface

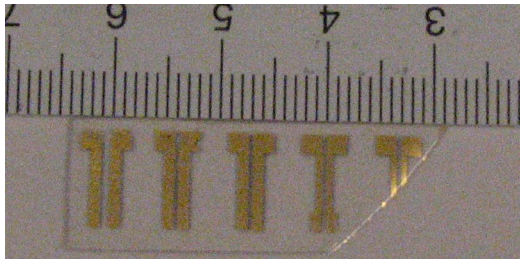

Fig. 4: Gold electrode sandwiched glucose sensors

Fig. 5. The fabricated PDMS based sensors do not lose their conductivity even though they are bent.

### *B. Results for the current response*

After successful fabrication, the current response of the glucose sensor is measured. The current response is measured by submerging the sensors in appropriate solution. First of all, the current generated in the high purity water using the gold only electrodes is measured. The current response (Fig. 6 and Fig. 7) is measured and logged using a potentiostat. The current drift in high purity water is plotted in Fig. 6 and response of the sensor with increasing $H_2O_2$ concentrations is shown in Fig. 7.

The current generated in high purity water is less than 10 nA which clearly indicates no drift in the sensor current (Fig. 6). The linearity test (Fig. 7) with increasing $H_2O_2$ concentration also indicates that the sensor current also increases with increasing $H_2O_2$ concentration.

## VI. CONCLUSION

A flexible polymer based electro-enzymatic glucose sensor using a new two layer PDMS fabrication process, SU-8 as a lift off layer, and gold electrodes is presented for the first time. The new fabrication process for flexible sensors with transparent hybrid polymer fabrication technique using PDMS and SU-8 is successfully demonstrated. The total thickness of the fabricated sensors is measured 700 µm. The actual sensor size in all the designs is 2mm x 3mm. Both active and reference gold electrodes are successfully tested. The PDMS polymer based glucose sensor is economical to fabricate, with the use of gold electrodes making the fabrication process very simple to perform in any basic micro-fabrication facility.

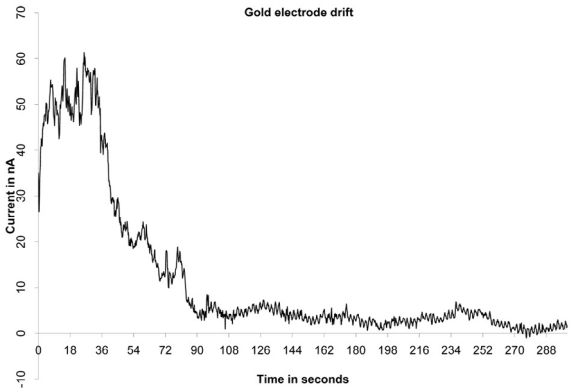


Fig. 6: Gold electrode current with high purity water

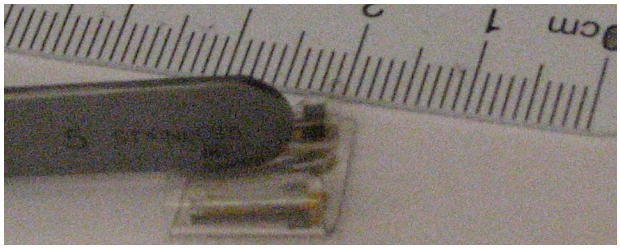

Fig. 5: Flexibility of the PDMS based sensor

Post fabrication tests such as electrical conductivity, profilometry, and $H_2O_2$ concentration response are utilized to characterize the process and working sensors. Optical imaging and electrical conductivity tests show for the first time successful fabrication of the electro-enzymatic sensor using the novel materials in the dual layer PDMS process with sandwiched gold electrodes. A linearity measurement is successfully done on working sensors, showing a linear response for sensors.

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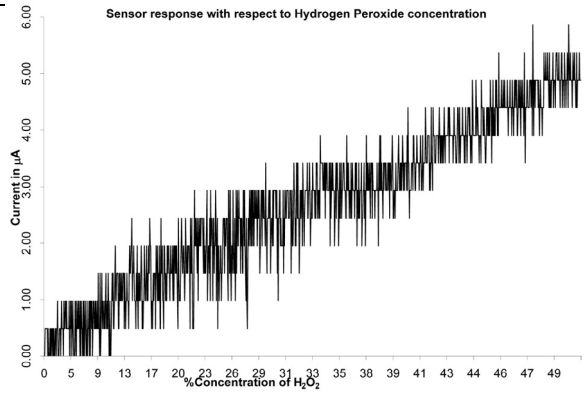


Fig. 7: Linearity response of the sensor with respect to $H_2O_2$ concentration.