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Chang-Soo Kim
Missouri University of Science and Technology, ckim@mst.edu

Jongwon Park

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INFLUENCE OF OXYGEN MICROENVIRONMENT ON MICROFLUIDIC GLUCOSE SENSOR PERFORMANCE

Chang-Soo Kim\textsuperscript{1,2}, Jongwon Park\textsuperscript{1}
\textsuperscript{1}Department of Electrical & Computer Engineering, University of Missouri-Rolla, MO, USA
\textsuperscript{2}Department of Biological Sciences, University of Missouri-Rolla, MO, USA

Abstract - We propose a novel method to overcome significant problems of baseline drift and sensitivity degradation in amperometric biosensors based on oxidase enzyme reactions. A novel glucose microsensor with a built-in electrochemical oxygen manipulation microsystem is introduced to demonstrate three novel functionalities: one-point in situ self-calibration (zero-point), broadening of dynamic range and increase in sensitivity. The influence of electrochemically generated oxygen microenvironment on the sensor output within a fluidic structure is investigated.

Keywords - electrolysis, bubble, oxygen, hydrogen

I. INTRODUCTION

Significant problems of most biosensors, including the glucose sensors, are unpredictable baseline drift and sensitivity degradation during continuous use. The sensors for long-term use should employ a dependable, simple, and convenient calibration procedure to verify accuracy and to check functionality. Another major limitation in the use of oxidase enzyme based biosensors is the dependence of enzyme activity on background oxygen concentrations. To overcome the latter problem, anaerobic operation of a glucose sensor with pulse techniques was proposed \cite{1}. The use of a diffusion membrane to increase the oxygen/glucose ratio \cite{2} and electrodes mediators to complete the enzyme reaction without oxygen \cite{3} is other examples to minimize this oxygen dependency of enzyme reactions. However, all these techniques still have the problems of baseline drift and sensitivity degradation during long-term monitoring.

This study proposes for the first time a novel glucose microsensor with an integrated electrochemical oxygen manipulation microsystem within a fluidic structure to achieve three novel functionalities simultaneously; (1) one-point in situ self-calibration (zero-point), (2) broadening of dynamic range, and (3) increase in sensitivity. The glucose oxidase (GOD) reaction is as follows;

\[ \beta-D\text{-Glucose} + O_2 \rightarrow \text{Gluconic acid} + H_2O_2 \]

The zero-point self-calibration can be done with a hydrogen bubble which is generated at the calibration electrode by water electrolysis. When the microsensor is surrounded by the hydrogen bubble, this oxygen-depleted environment does not allow the enzyme reaction to occur as in the reaction above, thus mimicking a glucose-free sample for a zero-point in situ calibration. The dynamic range and sensitivity of the sensor can be improved by providing a constant oxygen environment around the sensor with an oxygen bubble generated by the same water electrolysis.

II. EXPERIMENT

The microsystem in Fig. 1 is composed of an on-chip syringe, an on-chip calibrator, and a glucose microsensor. An electrochemical instrument was used to provide the chronamperometric operation for the glucose sensor and the galvanostatic operation for the water electrolysis actuation and calibration. A pair of calibration electrodes provides a pair of oxygen and hydrogen bubbles for the microsensor. To control this local environment surrounding the microsensor, another actuation bubble within an on-chip syringe was used to move these calibration bubbles along the microchannel. The generation and collapse of the actuation bubble give a bidirectional movement of the calibration bubbles. The size of the counter chamber is larger than that of the actuation chamber to avoid the generation of unnecessary hydrogen bubbles using the larger electrode in the counter chamber.

Fig. 2 shows the photograph of a fabricated fluidic system. A thick photosresist was used to prepare a template for the molding process of a poly-dimethylsiloxane (PDMS) cover layer to include the microchannel structure. The electrode substrate was a silicon wafer with a silicon nitride layer coating. A platinum/titanium thin film was deposited by e-beam evaporation and patterned by lift-off.
technique to define the electrodes. An amperometric sensor, based on H$_2$O$_2$ detection, was prepared by immobilizing GOD on the 2-electrode thin films.

III. RESULTS & DISCUSSION

Fig. 3 (a) is the dispensed volumes with respect to the actuation current duration of the on-chip syringe, which is directly proportional to the dislocation of the calibration bubbles along the microchannel. It shows the volume expansion of the oxygen actuation bubble to give excellent linear relationships with respect to the duration of the actuation current.

The sensor outputs were compared in Fig. 4 according the three different oxygen environments, that is, when the microsensor was covered by the hydrogen bubble, by the oxygen bubble, and by a normal aerated solution, respectively. The hydrogen bubble provided a zero-point calibration environment (i.e., 0% oxygen) for the microsensor, while the oxygen bubble provided a constant background oxygen environment (i.e., 100% oxygen). The outputs with the hydrogen bubble are virtually zero regardless of the actual glucose concentrations in the sample, since the necessary oxygen is not available from the surrounding environment. Also, a wider dynamic range and a higher sensitivity were obtained due to the constant background oxygen environment provided by the oxygen bubble.

IV. CONCLUSION

The controlled oxygen microenvironment by an on-chip microsystem effectively manipulated the responses of the fluidic glucose microsensor. This result suggests that the one-point (zero-point) calibration is possible with the hydrogen bubble, mimicking a glucose-free solution even in the presence of glucose in the sample. Also a wider dynamic range and a higher sensitivity are anticipated utilizing the oxygen bubble to ensure that the reaction is not perturbed by oxygen.

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