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High-Performance and Low-Cost Microfluidic-Electrohemical Biosensors

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Summary:
Microfluidic-electrochemical biosensors were fabricated by printing capillary microfluidic channels over printed electrodes using a functional material. The printed sensors without electrode modification were tested with a limit of detection (LOD) of 7 μM on glucose detection. This work has demonstrated the possibility of achieving high sensing performance with low-cost sensors, and reported how the printed microfluidic channels bring the performance to this level.

Keywords: Biosensors, microfluidic, electrochemical, printing.

Introduction

Microfluidic-electrochemical biosensors are normally fabricated by integrating microfluidic hollow channels with electrodes [1] or printing electrodes on paper-based microfluidic channels [2]. Both of them are not suitable for fabricating high-performance devices at low cost. Our invented capillary microfluidic technology [3] makes it possible to print the devices in volume at low cost. More importantly, the printed sensors are characterized with very high sensitivity without any surface modification of electrodes using expensive materials and processes. This paper presents the method of printing the sensors, the application of the sensors in glucose detection, and the effects of the microfluidic channels on sensing performance.

Experiment

Commercial Ag/AgCl inks (Kayaku Advanced Materials) and carbon inks (Loctite) were used to print reference, working and countering electrodes on transparent PET films, followed by the printing of microfluidic channels on their top using a functional material. The functional materials were formulated in house by mixing alumina particles, silica particles, polyvinyl alcohol and dimethyl sulfoxide (Millipore Sigma). An ASYS EKRA X1-SL semi-automatic screen printer (ASYS Group was used for the printing.

For the glucose detection using the printed electrochemical biosensors, a Princeton Applied Research PARSTAT 2263 potentiostat (AMETEK Scientific Instrument) was used for both cyclic voltammetry and chronoamperometry measurement. In In the analysis, 1 μL of enzyme (glucose oxidase) solution containing 100 mM potassium ferricyanide and 1 M KCl in 0.1 M PBS at pH 7.4 was first added onto each microfluidic channel over the working electrode and was allowed to dry at room temperature. Then, 6 μL of the glucose sample was added to the channel inlet and allowed to travel through the whole channel (within 1 minute). The experiments were performed between a potential of −0.3V and 1V and at scan rates ranging from 25 to 400 mV/s.

Results

Fig. 1 illustrates the process of printing microfluidic-electrochemical sensors using the method and materials. The obtained microfluidic channels are characterized with standalone structure and can transport fluid through capillary action in the similar fashion as the paper-based microfluidic channels do (Fig. 2).

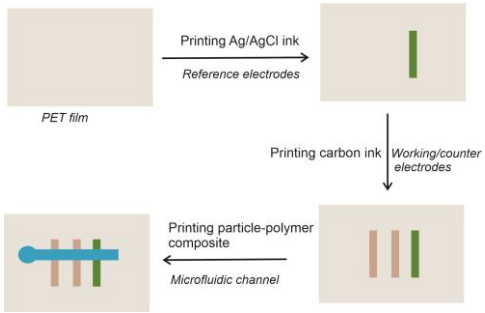

Fig. 1. Process flow of printing microfluidic-electrochemical biosensors.

Fig. 2. Photograph of printed straight capillary microfluidic channels. 2 seconds after a diluted inkjet ink was dropped onto the inlet of the central channel, and a colored paper was placed underneath for photo taking.

Fig. 3 shows the printed sensors with a single testing channel and multiple testing channels for enzymatic detection. After an enzyme solution is dopped onto the microfluidic section over the electrodes, testing samples can be dropped onto the circular inlets of the microfluidic channels for the detection. Many sensors can be printed on a transparent PET film with a high yield using a pilot-scaled screen printer, and the process can be implemented in sheet-to-sheet and even roll-to-roll for volume production.

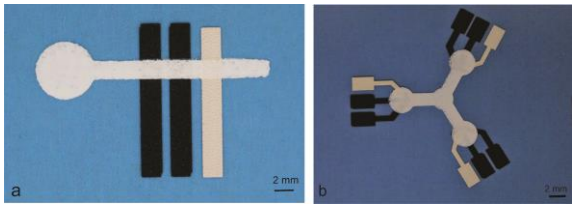

Fig. 3. Photographs of printed microfluidic-electrochemical biosensors for enzymatic detection. a, device for single reaction detection; b, device for simultaneous detection of three types of enzymatic reactions. The devices were printed with a screen printer, and colored papers were placed underneath for photo taking.

The printed microfluidic channels play a vital role in sensing performance through their porous structure. The interconnected pores, that are formed from the packing voids of the inorganic particles in the particle-polymer composite (Fig. 4), can substantially enhance the sensitivity while transporting testing samples to electrode surfaces through capillary action. This type of enhancement was not reported in any electrochemical sensors. A very lower polymer concentration in the composite was found in favor of the enhancement effect but mechanically weakening the printed structure. A two-layer microfluidic channel structure was thus developed by first printing the composite with 3% polymer on the electrodes for high sensing performance and then printing the composite with 8% polymer on top to provide mechanical protection. As such, sensors with high sensing performance and sufficient mechanical strength can be printed in large quantity.

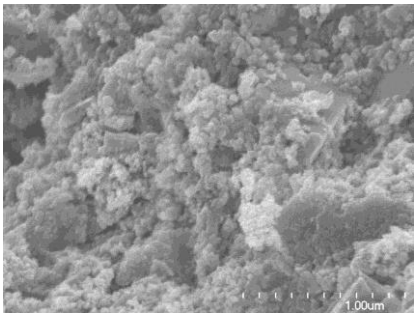

Fig.4. SEM image of the microfluidic channel of one printed biosensor. The particle-polymer composite composing of 97% alumina and silica particles, and 3% polyvinyl alcohol was printed with a screen printer.

The printed biosensors were tested in glucose detection (Fig. 5) with a limit of detection (LOD) at 7 μM obtained, due to the powerful sensitivity enhancement by the pores in microfluidic channels. A LOD in such low level was only reported when expensive nano materials with complicated processes were used in fabricating electrochemical biosensors. This work has demonstrated the possibility of fabricating high-performance biosensors at low cost.

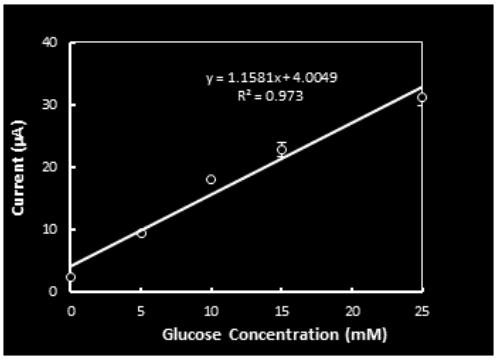

Fig. 5. Chronoamperometric calibration curve for glucose concentration as a function of current at 30 s. The biosensors with a structure in Figure 3a were printed with the composite with 3% PVA on bottom and the composite with 8% PVA on top.

References

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