



Development of a disposable glucose biosensor using electroless-plated Au/Ni/copper low electrical resistance electrodes

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ABSTRACT

This paper presents a glucose biosensor, which was developed using a Au/Ni/copper electrode. Until now, research regarding the low electrical resistance and uniformity of this biosensor electrode has not been conducted. Glucose oxidase (GOD) immobilized on the electrode effectively plays the role of an electron shuttle, and allows glucose to be detected at 0.055 V with a dramatically reduced resistance to easily oxidizable constituents. The Au/Ni/copper electrode has a low electrical resistance, which is less than 0.01 Ω , and it may be possible to mass produce the biosensor electrode with a uniform electrical resistance. The low electrical resistance has the advantage in that the redox peak occurs at a low applied potential. Using a low operating potential (0.055 V), the GOD/Au/Ni/copper structure creates a good sensitivity to detect glucose, and efficiently excludes interferences from common coexisting substances. The GOD/Au/Ni/copper sensor exhibits a relatively short response time (about 3 s), and a sensitivity of 0.85 $\mu\text{A mM}^{-1}$ with a linear range of buffer to 33 mM of glucose. The sensor has excellent reproducibility with a correlation coefficient of 0.9989 ($n = 100$ times) and a total non-linearity error of 3.17%.

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1. Introduction

Glucose investigation is very important in numerous situations. It is of importance in the food industry for quality control purposes, in fermentation and, most importantly, as a clinical indicator of diabetes (American Diabetes Association, 1994, 1999). In addition, self-monitoring of blood glucose is a critical aspect of diabetes care (Owen, 1995; Nakamura and Karube, 2003; Newman and Turner, 2005). The requirements for *in vitro* glucose monitoring systems to measure glucose concentrations in capillary blood samples as well as procedures to verify and validate their performance by the intended users are specified by international standard 15197 (ISO 15197 First edition, 2003). These systems are intended for self-testing by laypersons to manage diabetes mellitus. International Standard 15197 is applicable to manufacturers of such systems, while other organizations (e.g. regulatory authorities and conformity assessment bodies) are responsible for assessing the performance of these systems. The glucose meter is a medical device used to determine the approximate concentration of glucose in the blood. A small drop of blood, obtained by pricking the skin with a lancet, is placed on a disposable test strip, which the meter

reads and uses to calculate the blood glucose level. The meter then displays the level in mg dL^{-1} or mmol L^{-1} (Podczasy et al., 1997). Electrochemical glucose sensors based on an electrode modified with immobilized glucose oxidase have been reported previously (Sulak et al., 2006). A disposable amperometric biosensor to detect blood glucose has also been developed. Various biosensor electrode fabrication techniques have been reported in the literature, including rolling, screen printing (Crouch et al., 2005; Guan et al., 2005), and sputter deposition techniques (Hashimoto et al., 2007). Typically, a biosensor is based on a screen printed electrode. A carbon-paste electrode (CPE) is made from a mixture of conducting graphite powder and a pasting liquid. These electrodes are simple to make, and offer an easily renewable surface for electron exchange. These electrodes are widely used, mainly for voltammetric measurements. Carbon paste-based sensors (Yabuki et al., 1992; Ge et al., 1998; Boujtita et al., 2000; Hong et al., 2002; Hart et al., 2002; Fährnich et al., 2003; Darain et al., 2003; Gao et al., 2003; Honeychurch et al., 2003; Darain et al., 2005; Díaz-González et al., 2005; Ledru et al., 2006; Corgier et al., 2007; Piermarini et al., 2007; Tangkuaram et al., 2007; Honeychurch et al., 2007; Djellouli et al., 2007) are also applicable in coulometry (both amperometry and potentiometry). The wide application of a carbon paste as the matrix of an enzyme electrode is attributed to the simple preparation procedure, which facilitates mass production and therefore, is economical. Electrodes made in this way are highly selective

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sensors for both inorganic and organic electrochemistry. However, in practice, printing conditions must be adjusted to control thickness as well as electrode area, which are known to proportionally affect the sensor response. Screen printing consists of a screen, a squeegee, and ink, and is a type of stencil printing. A screen is a type of mesh, which has holes that are clogged by emulsion to leave the desired patterns. A squeegee is a rubber spatula, which squeezes ink out of screen's holes onto a base film (Kaimori et al., 2006).

With regard to the screen printing method, many factors are known to affect the accuracy of printing.

- A. Screen printer: printer type, print speed, clearance, and print pressure.
- B. Squeegee: squeegee type, hardness, attack angle.
- C. Screen: frame size, print area, print pattern, gauze, thickness, opening, mesh material, the number of meshes, emulsion material, emulsion thickness.
- D. Ink: ink type, viscosity, thixotropy, leveling, stirring, wettability to base film and other ink layers, drying method.
- E. Base film: affinity to ink, heat contraction.

To minimize the dispersion of the electrode area, the print speed, clearance, print pressure, squeegee hardness, attack angle, print area, print pattern, mesh material, the number of meshes, ink type, thixotropy, and heat contraction are all important factors in adjusting the screen printing conditions. Nevertheless, until now, research has not been conducted on the low electrical resistance and uniformity of this biosensor electrode. Many of the issues associated with glucose biosensors have very little to do with the device itself. In virtually every case, the basic design of commercially successful biosensors has not changed significantly. Herein we report a biosensor solution to this electrical resistance. In this work, we have developed a new form of an enzyme electrode to improve the performance of disposable glucose sensors. We fabricated electrodes with an Au/Ni/copper structure, which offer a very low uniform electrical resistance of about $0.01\ \Omega$, by attaching a copper film to a plastic film substrate using a laminating method and subsequent plating of nickel and gold on to it with an electroless technique. The fabrication of the enzyme electrode was completed by immobilizing the enzyme on the electrode surface. The fabricated glucose sensor exhibits a very rapid sensing time (3 s) with an excellent reproducibility and fabrication yield.

2. Experimental

2.1. Materials

All structural materials of the sensor chip and reagents were commercially available: glucose oxidase (GOD, EC 1.1.3.4, 15,000–25,000 units g^{-1} , Type II from *Aspergillus niger*), analytical-reagent grade potassium hexacyanoferrate (II) (i.e. ferricyanide) with 99.9% purity. Triton X-100 wetting agent and β -D-(+)-glucose were purchased from Sigma-Aldrich. Deionized (D.I.) water was used throughout the experiments to prepare the samples, buffers, and enzyme solutions. A 0.1 M phosphate buffer solution (PBS) was freshly prepared by mixing solutions of KH_2PO_4 and K_2HPO_4 . The pH of the prepared solution was 7.03. Enzyme solutions were prepared by dissolving solids in the above-mentioned buffer solutions. The 0.1 M of GOD solutions were immobilized on the Au/Ni/copper electrode using the physical absorption method. To prepare the enzyme electrode, 0.5 μL of the enzyme solution (1.2 g of GOD in 10 mL of PBS) was dropped onto the sensing area with a 5 mm² channel size using syringe pump. Then, 0.5 μL of the electron transfer mediator (0.1 M potassium ferricyanide solution) was also

immobilized onto the sensing area using the same method. The sensor can be measured after drying for 24 h at room temperature. 0.1 M sample stock glucose solution was prepared daily, and stored overnight to reach mutarotational equilibrium before use.

2.2. Apparatus

In this study, electrochemical measurements to estimate the newly developed sensor chips, which consist of a two electrode system, were performed using an electrochemical analyzer. The equipment employed included a cyclic voltammeter, an amperometer (WonATech Co., Ltd), and WPCIPG software version 1.0 installed on a personal computer. The Au/Ni/copper electrodes on polyethylene terephthalate (PET) substrates were custom made at Jaein Circuit Co., Ltd, Korea. All electrochemical experiments were carried out in a cell containing 0.25 μL 0.1 M phosphate buffer solution (PBS, pH 7.03) at room temperature ($25 \pm 2^\circ\text{C}$) using Au/Ni/copper as the working and reference electrodes.

2.3. Measurement method

In this paper, we used cyclic voltammetry and amperometry techniques to study the electrochemical properties of the immobilized GOD on Au/Ni/copper electrodes for biosensor applications. GOD and potassium hexacyanoferrate (II) can be immobilized by simple adsorption on a Au/Ni/copper electrode. In addition, the ferro-ferri cyanide redox coupling electrochemical reaction (Cass et al., 1984) was used as the sensing mechanism for this glucose sensor. The ferro-ferri redox reaction can occur at a GOD/Au/Ni/copper electrode surface, which produces an oxidation and a reduction current from the redox reaction. However, GOD cannot directly transfer electrons to conventional electrodes in amperometric sensors because its active site is embedded within the enzyme layer. In this work, potassium ferrocyanide was used as the mediator (Dong et al., 1992; Pasco et al., 2004). The fabricated glucose biosensor was based on the oxidation of glucose according to the following reactions. Fig. 1 shows the proposed mechanism for GOD/Au/Ni/copper to be oxidized by the entrapped GOD at a polarization of 0.055 V. Potassium ferricyanide had an electronic mediator function, and received electrons to become potassium ferrocyanide. Simultaneously, glucose was oxidized with gluconolactone ($\text{C}_6\text{H}_{10}\text{O}_6$). Ultimately, the electric current (e^-) was detected by the Au/Ni/copper electrode, and potassium ferrocyanide changed to potassium ferricyanide.

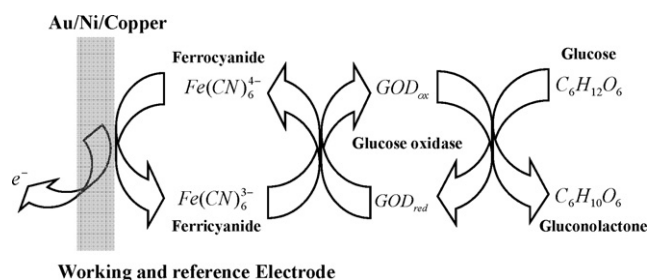
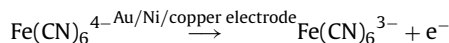
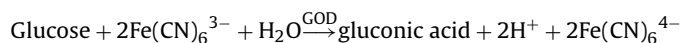


Fig. 1. Proposed reaction mechanism of the GOD/Au/Ni/copper glucose sensor polarized at 0.055 V vs. CV (cyclic voltammograms).

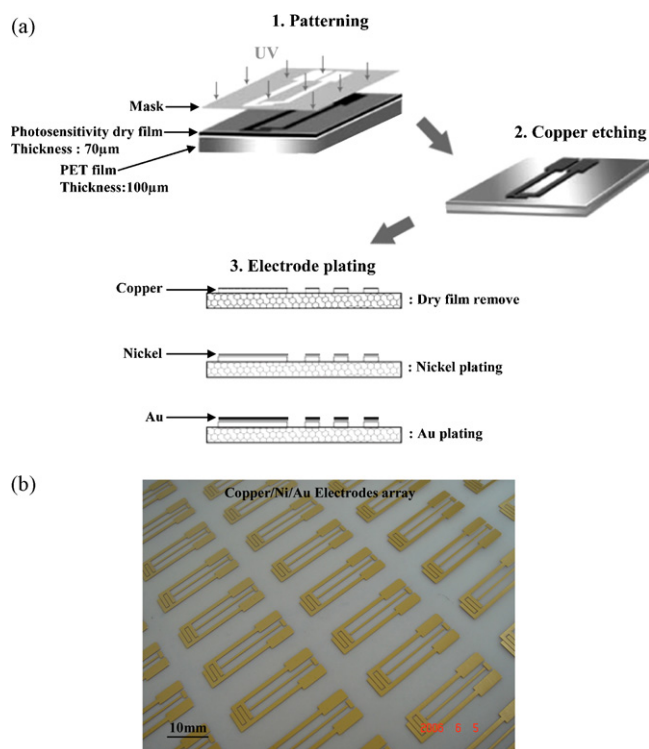


Fig. 2. (a) Manufacturing method for the Au/Ni/copper electrodes. (b) Photograph of an array of fabricated Au/Ni/copper electrodes.

2.4. Au/Ni/copper electrodes

The Au/Ni/copper electrodes were fabricated using heat to laminate a 20 μm copper film on to a 100 μm thick PET substrate. Then an ultraviolet (UV) photosensitive dry film was adhered using the laminating method as shown in Fig. 2(a). A reverse photo mask was used to create the copper electrode pattern by UV exposure and wet etching. The copper electrode is a metal, which does not react with water (H₂O), but the oxygen in the air will react slowly at room temperature to form a layer of copper oxide on the copper metal. Nickel plating was used in order to improve the plating adhesion characteristics of gold and copper. Gold plating is often used in electronics to provide a corrosion-resistant, electrically conductive layer on copper, typically in electrical connectors and printed circuit boards. Moreover, the surface of gold plating was stabilized electrochemically, and the immobilization of the enzyme was easier. In this study, the copper electrode was followed by sequential electroless-plating of nickel and gold. Fig. 2(b) shows a photograph of the fabricated Au/Ni/copper electrodes, while Fig. 3(a) and (b) are a schematic of the disposable glucose biosensor and a photograph, respectively. The glucose biosensor consisted of a bottom, a middle, and upper plate. The fabrication process of the sensor is as follows:

- The working and reference electrodes are formed on the fabricated bottom plate.
- Then the bottom plate is combined with the middle plate.
- Enzyme solution (GOD + hexacyanoferrate II) is deposited on the channel of the combined structure (formed bottom plate + middle plate).
- Holes to form the inlet and outlet are fabricated in the upper plate.
- The glucose biosensor is accomplished by a combination between (c) and (d).

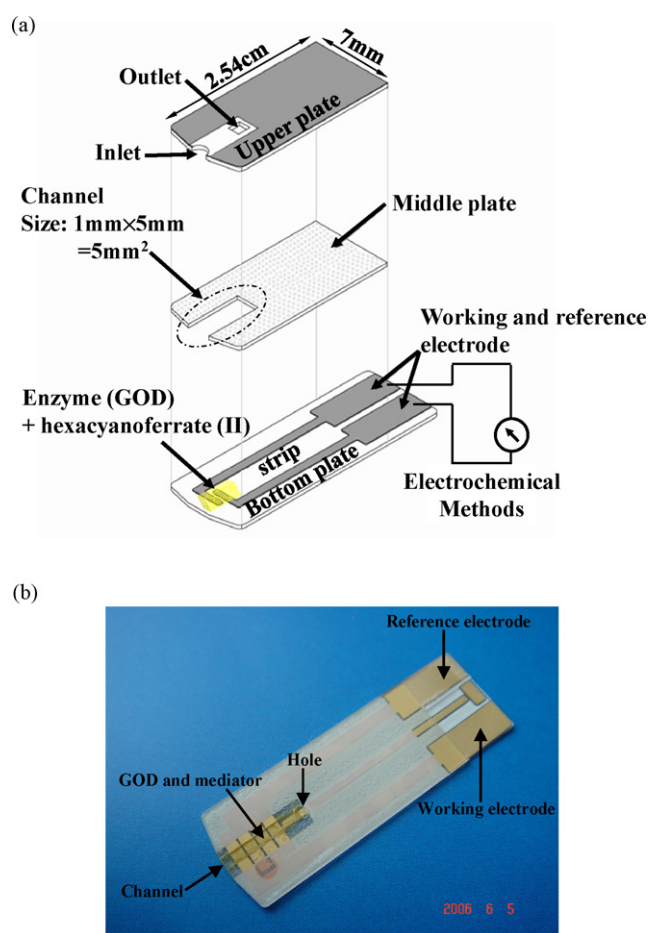


Fig. 3. (a) Schematic of the biosensor. (b) Configuration of the GOD/Au/Ni/copper disposable glucose biosensor.

- The resistance of the fabricated Au/Ni/copper electrode was about 0.01 Ω. This low resistance offered a lower operating voltage.

3. Results and discussion

3.1. Cyclic voltammograms of glucose solutions

Glucose solutions with different buffer concentrations, 1.1, 2.5, 5.5, 8.5, 11.5, 13.5, 16, 21, 25, and 33 mM, were prepared prior to conducting electrochemical measurements. After adding the enzyme GOD and the mediator, cyclic voltammetry was conducted for each concentration of glucose solution, and the response current was recorded. The CV results obtained at each concentration of glucose, glucose oxidase, and mediator showed that the response current increased in proportion to the glucose concentration in the specific potential range (i.e., around +0.055 V for the Au/Ni/copper electrode oxidation peak and around −0.055 V for the Au/Ni/copper electrode reduction peak). However, in this study, the 20 μm copper film on a PET film formed a biosensor electrode with a very low electrical resistance. Then the biosensor electrode was fabricated using an electroless-plating method to form a 5 μm thick gold and nickel films on the fabricated copper/PET film. The fabricated electrode surface consisted of the electroless-plated gold, which was electrochemically stable. The Au/Ni/copper/PET electrode had a low electrical resistance, and it is possible to mass produce the biosensor electrode with a uniform electrical resistance. The low electrical resistance has an advantage in that the redox peak occurs

at low voltage. Fig. 4(a) and (b) shows the CV results obtained (using scan rates of 50 mV/s). Glucose was oxidized by the electrochemical transformation of potassium ferricyanide to potassium ferrocyanide. The potassium ferrocyanide was continuously regenerated by the electrode under oxidizing conditions. The size of the response current depended on the enzyme and glucose concentrations. The electro-catalytic activity of the Au/Ni/copper electrode towards the oxidation of glucose solution was demonstrated. In the glucose solutions, the cyclic voltammetry measurements showed the characteristics of the Au/Ni/copper electrode, including the oxidation (−0.9 V to +0.9 V) and reduction (+0.9 V to −0.9 V) regions. Moreover, it showed symmetrical oxidation peaks and reduction peaks for glucose concentrations from the buffer to 33 mM. The CV characteristic of the glucose redox reaction was generated in the 5 mm² Au/Ni/copper sensing area.

3.2. Current–time relationship to measure glucose in solution

Glucose was detected by the electrochemical biosensors based on the electrochemical oxidation of hydrogen peroxide generated by the enzyme-catalyzed oxidation of glucose at anodic potentials (+0.6 V vs. Ag/AgCl) (Ianniello and Yacynych, 1981; Wang et al., 1992). However, at this relatively high potential, other oxidizable species such as ascorbic acid, uric acid, and acetaminophen may interfere with the measurement. To avoid interference, some improved biosensors have been used to detect hydrogen peroxide at low potential (Yang et al., 2006; Sun et al., 2006; Crespiho et al., 2006). The low electrical resistance has the advantage that the redox peak occurs at low voltage. Fig. 4(b) and (c) shows the response current–time relationship for glucose solutions of different concentrations using Au/Ni/copper working and reference electrodes. The response currents were obtained from the CV results (at +0.055 V) conducted at each glucose concentration after injecting the glucose solutions with the enzyme GOD and a mediator. As shown in Fig. 4(c), the elapsed time to reach steady state (constant current) was relatively short for each glucose concentration. Because the screen printed carbon electrode had a higher electric resistivity (approximately 80–1 k Ω $\pm$ 5–10%) than the Au/Ni/copper electrode (about 0.01 Ω), the screen printed carbon electrode required a high applied potential (approximately 0.2–0.7 V). Therefore, the enzymatic reaction will require an increased response time to reach a steady state (equilibrium). Because the fabricated electrode can reach a steady state within 3 s, it has a quicker response time than the screen printed carbon electrode. The results clearly demonstrate that the response is very fast, which implies that the sensor will be efficient in detecting glucose.

3.3. Reproducibility with linearity of the Au/Ni/copper electrode glucose biosensor

The reproducibility of the fabricated sensor was assessed. The sensor was tested using the amperometric method. A calibration

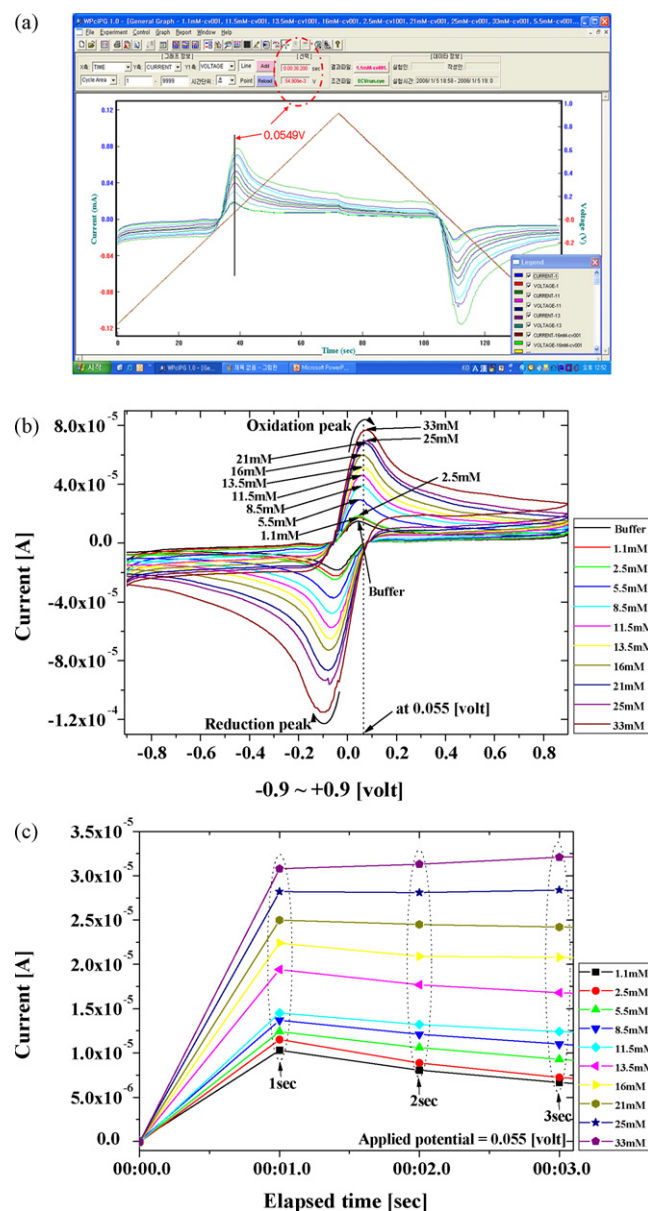


Fig. 4. (a) Results of cyclic voltammetry (WonATech Co., Ltd) and (b) cyclic voltammetry results showing the current responses for each glucose concentration. Scan rate: 50 mV/s. Glucose solutions were prepared using a 0.1 M phosphate buffer (pH 7.0). (c) Amperometric profiles of the disposable glucose biosensor showing the change in the response current of each glucose solution with time. Working and reference electrodes are Au/Ni/copper. Response currents are obtained from the CV results. Applied potential is +0.055 V.

Table 1
Repeatability evaluation of different glucose concentrations

Glucose concentration, mmol L ⁻¹ (mg dL ⁻¹), N = 100	Average (A), $m = (1/n) \sum x_i$	Relative standard deviation (A), $\sigma^2 = (1/n) \sum (x_i - m)^2$	Coefficient of variation (%), (standard deviation/arithmetic average) $\times 100$
1.7–2.8 (30–50)	7.37E–06	1.09697E–07	1.49
2.9–6.1 (51–100)	1.03E–05	5.70455E–07	5.54
6.2–6.1 (111–150)	1.26E–05	4.39065E–07	3.48
8.4–13.9 (151–250)	1.69E–05	6.82113E–07	4.04
14–22.2 (251–400)	2.45E–05	4.48454E–07	1.83
22.3–33 (401–594)	3.25E–05	8.544E–07	2.63
			Total non-linear error (CV) = 3.17 (%)

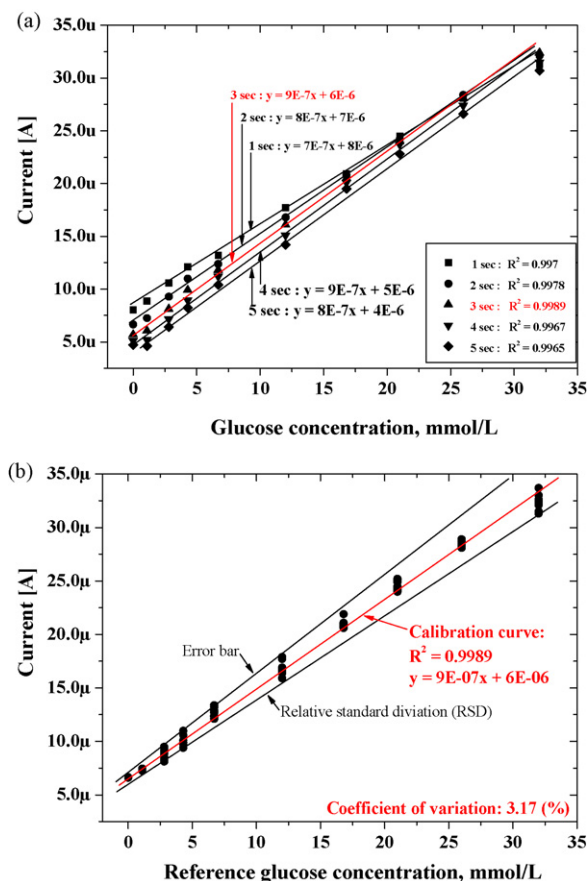


Fig. 5. (a) Characteristics of the linear correlation coefficient (R^2) as a function of time. 1 s: $R^2 = 0.997$, 2 s: $R^2 = 0.9978$, 3 s: $R^2 = 0.9989$, 4 s: $R^2 = 0.9967$ and 5 s: $R^2 = 0.9965$. (b) Repeatability of the Au/Ni/copper electrode glucose biosensor. Calibration curve: $R^2 = 0.9989$, $y = 9E-07x + 6E-06$.

curve was obtained from the amperometric response of the sensor at measurement times from 1 to 5 s. The Au/Ni/copper electrode achieved a steady state current in less than 3 s with a linear correlation coefficient (R^2) of 0.9989, as shown in Fig. 5(a). Fig. 5(b) shows the characteristic measurement of the fabricated disposable (Au/Ni/copper electrode) glucose biosensor, which was repeated 100 times, and indicates that the disposable (Au/Ni/copper electrode) glucose biosensor has good linearity and considerable reproducibility. In Table 1, the total non-linearity error is 3.17% (American Diabetes Association, 1999; Clarke et al., 1987). According to ISO 15197 (ISO 15197 First edition, 2003), the permitted production standard for a blood glucose sensor for clinical applications is a non-linear error ratio within $\pm 7\%$.

4. Conclusion

A disposable glucose biosensor with improved sensor characteristics is fabricated. The glucose biosensor is comprised of an Au/Ni/copper electrode structure on a PET substrate. The fabricated electrode has a low electrical resistance (0.01Ω). Because of the low electrical resistance of the electrode, the sensor can be used with a low operating potential (0.055 V) while maintaining a good sensitivity to detect glucose and efficiently exclude interference from common coexisting substances. Thus, the Au/Ni/copper electrode sensor has a wide linear response range for glucose concentrations of $1.0\text{--}33.0 \text{ mM}$ and a high sensitivity, with a linear correlation coef-

ficient of 0.9989. Furthermore, the clinical minimum blood demand quantity for measurement is only $0.25 \mu\text{L}$. Future studies include using this fabricated sensor to measure interference from ascorbic acid, uric acid, and acetaminophen in real blood samples. Hence, it is anticipated that the Au/Ni/copper electrode will be a neo-disposable biosensor without compensation (no coding type) in the healthcare field because the fabricated electrode may be mass produced with a uniform and low electrical resistance.

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References

- American Diabetes Association, 1994. *Diab. Care* 17, 81–86.
- American Diabetes Association, 1999. *Diab. Care* 22, S77–S78.
- Boujtita, M., Hart, J.P., Pittson, R., 2000. *Biosens. Bioelectron.* 15 (5–6), 257–263.
- Cass, A.E.G., Francis, D.G., Hill, H.A.O., Aston, W.J., Higgins, I.J., Plotkin, E.V., Scott, L.D.L., Turner, A.P.F., 1984. *Anal. Chem.* 56, 667–671.
- Clarke, W.L., Cox, D., Gonder-Frederick, L.A., Carter, W., Pohl, S., 1987. *Diab. Care* 10, 622–628.
- Corgier, B.P., Marquette, C.A., Blum, L.J., 2007. *Biosens. Bioelectron.* 22 (7), 1522–1526.
- Crespilho, F.N., Ghica, M.E., Florescu, M., Nart, F.C., Oliveira Jr., O.N., Brett, C.M.A., 2006. *Electrochem. Commun.* 8 (10), 1665–1670.
- Crouch, E., Cowell, D.C., Hoskins, S., Pittson, R.W., Hart, J.P., 2005. *Biosens. Bioelectron.* 21 (5), 712–718.
- Darain, F., Park, D.S., Park, J.S., Chang, S.-C., Shim, Y.-B., 2005. *Biosens. Bioelectron.* 20 (9), 1780–1787.
- Darain, F., Park, S.-U., Shim, Y.-B., 2003. *Biosens. Bioelectron.* 18 (5–6), 773–780.
- Díaz-González, M., González-García, M.B., Costa-García, A., 2005. *Biosens. Bioelectron.* 20 (10), 2035–2043.
- Djelloul, N., Rochelet-Dequaire, M., Limoges, B., Druet, M., Brossier, P., 2007. *Biosens. Bioelectron.* 22 (12), 2906–2913.
- Dong, S.J., Wang, B.X., Liu, B.F., 1992. *Biosens. Bioelectron.* 7 (3), 215–222.
- Fährnich, K.A., Pravda, M., Guilbault, G.G., 2003. *Biosens. Bioelectron.* 18 (1), 73–82.
- Gao, Q., Cui, X., Yang, F., Ma, Y., Yang, X., 2003. *Biosens. Bioelectron.* 19 (3), 277–282.
- Ge, F., Zhang, X.-E., Zhang, Z.-P., Xiao-Mei, Z., 1998. *Biosens. Bioelectron.* 13 (3–4), 333–339.
- Guan, W.J., Li, Y., Chen, Y.Q., Zhang, X.B., Hu, G.Q., 2005. *Biosens. Bioelectron.* 21, 508–512.
- Hart, J.P., Abass, A.K., Cowell, D., 2002. *Biosens. Bioelectron.* 17 (5), 389–394.
- Hashimoto, M., Sakamoto, N., Upadhyay, S., Fukuda, J., Suzuki, H., 2007. *Biosens. Bioelectron.* 22 (12), 3154–3160.
- Honeychurch, K.C., Hart, J.P., Pritchard, P.R.J., Hawkins, S.J., Ratcliffe, N.M., 2003. *Biosens. Bioelectron.* 19 (4), 305–312.
- Honeychurch, K.C., O'Donovan, M.R., Hart, J.P., 2007. *Biosens. Bioelectron.* 22 (9–10), 2057–2064.
- Hong, M.-Y., Chang, J.-Y., Yoon, H.C., Kim, H.-S., 2002. *Biosens. Bioelectron.* 17 (1–2), 13–18.
- Ianniello, R.M., Yacinyeh, A.M., 1981. *Anal. Chem.* 53, 2090–2095.
- ISO 15197, 2003. <http://www.iso.org/iso/home.htm>.
- Kaimori, S., Kitamura, T., Ichino, M., Hosoya, T., Kurusu, F., Ishikawa, T., Nakamura, H., Gotoh, M., Karube, I., 2006. *Anal. Chim. Acta* 573–574, 104–109.
- Ledru, S., Ruillé, N., Boujtita, M., 2006. *Biosens. Bioelectron.* 21 (8), 1591–1598.
- Nakamura, H., Karube, I., 2003. *Anal. Bioanal. Chem.* 377 (3), 466–468.
- Newman, J.D., Turner, A.P.F., 2005. *Biosens. Bioelectron.* 20 (12), 2435–2453.
- Owen, V.M., 1995. *Biosens. Bioelectron.* 10 (3–4), iv–v.
- Pasco, N., Baronian, K., Jeffries, C., Webber, J., Hay, J., 2004. *Biosens. Bioelectron.* 20 (3), 524–532.
- Piermarini, S., Micheli, L., Ammida, N.H.S., Palleschi, G., Moscone, D., 2007. *Biosens. Bioelectron.* 22 (7), 1434–1440.
- Podczasy, J.J., Flodstrom, G., Doucette, L., 1997. *Biosens. Bioelectron.* 12 (11), iii.
- Sulak, M.T., Gökdoğan, Ö., Gülce, A., Gülce, H., 2006. *Biosens. Bioelectron.* 21 (9), 1719–1726.
- Sun, Y., Yan, F., Yang, W., Sun, C., 2006. *Biomaterials* 27, 4042–4049.
- Tangkuaram, T., Ponchio, C., Kangkasomboon, T., Katikawong, P., Veerasai, W., 2007. *Biosens. Bioelectron.* 22 (9–10), 2071–2078.
- Wang, J., Naser, L., Angles, L., Hui, W., Chen, L., 1992. *Anal. Chem.* 64, 1285–1288.
- Yabuki, S., Mizutani, F., Katsura, T., 1992. *Biosens. Bioelectron.* 7 (10), 695–700.
- Yang, M., Yang, Y., Liu, Y., Shen, G., Yu, R., 2006. *Biosens. Bioelectron.* 21, 1125–1131.