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Ultra-sensitive biosensor with capacitive coupling-gate InGaZnO-based FET

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Abstract

In this study, an ultra-sensitive glucose sensing is achieved by capacitive coupling with an InGaZnO-based field effect transistor (FET). An insulated-gate InGaZnO-based FET shows the standard pH response of 56 mV/pH near the Nernst response because of no capacitive coupling-gate structure, while the pH sensitivity is enhanced to 313 mV/pH over the Nernst limit by using an extended-gate InGaZnO-based FET, the structure of which allows the capacitive coupling circuit. Using the extended-gate InGaZnO-based FET, the glucose response is about seven times higher, furthermore the remarkable detection limit of 10 nM was obtained according to capacitive coupling of the extended electrode with the oxide gate of FET. A platform based on the capacitive coupling-gate InGaZnO-based FET is suitable for a ultra-sensitive flexible device to monitor glucose levels in sweat on a skin in the future.

Keywords: InGaZnO-based biosensor, extended-gate FET, capacitive coupling, Nernst limit, and glucose

Introduction

Glucose sensors are widely used to monitor glucose levels in blood for diabetic mellitus patients. However, a conventional glucose sensor has several disadvantages; commercially available glucose sensors need blood sampling, which is accompanied by pain to measure blood glucose, and their detection sensitivity is insufficient at low glucose levels in biological fluids such as sweat, which is focused on as one of the samples for noninvasive glucose measurement instead of blood samples. Actually, the glucose levels in sweat are about one-tenth to one-hundredth of those in blood [1]. Therefore, the development of a novel glucose sensor, which enables to monitor low glucose levels in sweat, is required by use of flexible devices on a skin. In this paper, we focus on a thin film transistor (TFT) based on an amorphous-In-Ga-Zn-oxide (a- InGaZnO) for a flexible glucose sensor. In particular, we report the enhancement of pH and glucose sensitivities using the capacitive-coupling-gate InGaZnO-based FET.

Methods

As one of the InGaZnO-based FET, an insulated-gate FET was prepared, the insulated gate electrode of which was filmed on a semiconductor and common as the sensing electrode in a solution, as shown in **Fig. 1(a)**. The thicknesses of InGaZnO, ITO and SiO_2 comprising the gate insulator were 51 nm, 75 nm and 13 nm, respectively. The width (W) and length (L) of the gate channel, which was fabricated using a lithography technology, were 340 μm and 10 μm, respectively. Moreover, an extended-gate FET was arranged, a sensing electrode of which was separated from the IGZO-based FET, as shown in **Fig. 1(b)**. A change in charge density on the extended electrode with a fixed area is induced by pH variation and biological reactions, when the gate size of FET only can be varied. The size (d; diameter) of extended electrode with Ta_2O_5 or Au(Cr)/Ta_2O_5 was designed and fixed to be 18 mm, then the gate size of FETs, W (μm)/L (μm) was varied to be 50/50, 50/20, 50/10, 10/10 and 10/5, resulting in capacitance coupling of an extended electrode with an oxide gate of FET (**Fig. 2**)

Results and Discussion

Figure 3 shows the V_G-I_D electrical characteristics obtained in the solutions with various pH values using the insulated-gate InGaZnO-based FET (**Fig. 1(a)**). The threshold voltage, V_T shifted in the positive direction when the pH was increased from pH 4.01 to 9.18 at a constant I_D. This positive shift indicates a decrease of positive charges based on H^+, because the Ta_2O_5 gate surface is covered by hydroxyl groups in the solutions, which are sensitive to H^+. The pH response of the oxidized gate surface should be Nernstian (59.1 mV/pH at 25 °C). Actually, ΔV_T was about 56 mV/pH. Similarly, the change in the gate surface potential ($-\Delta V_T$) with the pH was investigated in a real-time manner, as shown in **Fig. 4**.

On the other hand, an extended-gate FET was utilized, a sensing electrode of which was separated from the InGaZnO-based FET (**Figs. 1(b)** and **5**). A change in charge density on the extended electrode (Ta_2O_5) with a fixed area (d=18 mm) was induced by pH variation, when the gate size (W/L) of FET only was varied. As a result, ΔV_T was induced to be approximately 198 mV/pH depending on pH using the extended-gate FET with W/L=10/10, as shown in **Fig. 6**. Moreover, the pH sensitivity was enhanced to 313 mV/pH over the Nernst limit at W/L=10/5 by changing the reciprocal of channel area (S based on W/L), 1/S (1/μm^2), from 4 x 10^{-4} to 2 x 10^{-2}, as shown in **Fig. 7**. This resulted from capacitive coupling (**Fig. 2**) and demonstrated the largest signal, compared with the previous data [2-4]. The enhancement of pH responsivity by the capacitive coupling method should contribute to the improvement of detection sensitivity for bio-molecules such as glucose.

In addition, the glucose response was enhanced by capacitive coupling using the InGaZnO-based FET with the extended electrode (Au(Cr)/Ta_2O_5) (**Figs. 1(b)**, **2** and **5**), as shown in **Fig. 8**. Actually, the electrical signal of the extended-gate FET with W/L=50/10 was about seven times higher than that obtained with the insulated-gate FET [5,6]. The signal, $|\Delta V_T|$ was induced by adding 1 mM of glucose, which was based on oxidation reaction of glucose at the Au surface. In particular, the capacitive coupling-gate glucose transistor showed a detection limit at lower concentration of glucose. That is, the remarkable detection limit of 10 nM was expected (**Fig. 9**). This means the glucose responsivity was improved three orders higher by the capacitive coupling method, which enables to detect even low glucose levels in sweat (**Fig. 9**).

Conclusions

A platform based on the capacitive coupling-gate InGaZnO-based FET is suitable for an ultra-sensitive biosensing system, which contributes to finding very small amount of bio-markers as well as glucose, in the field of pharmaceutical discovery, clinical diagnosis and environmental monitoring.

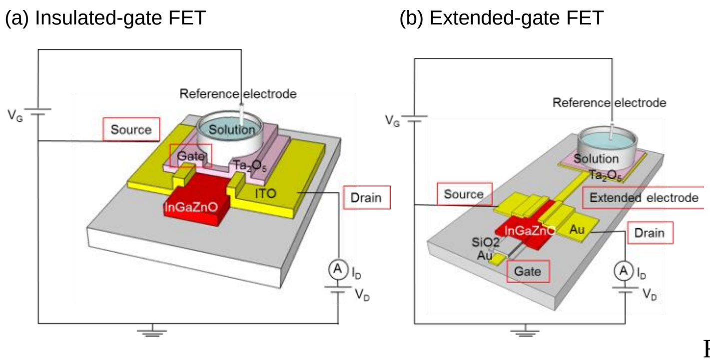


Fig. 1 Two types of InGaZnO-based biosensor. (a) Insulated-gate FET, (b) Extended-gate FET

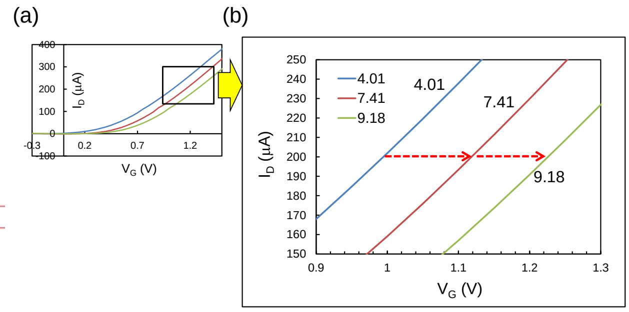


Fig. 3 V_G-I_D electrical characteristics of InGaZnO-based FET in various solutions. The pH was varied from 4.01 to 9.18. (b) is magnified graph of part of (a).

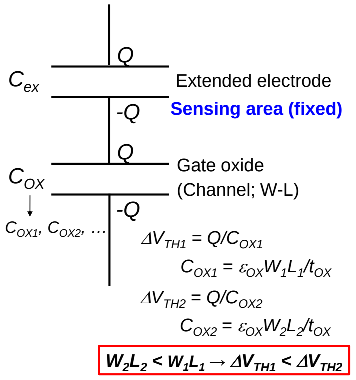


Fig. 2 Capacitive coupling with extended-gate FET.

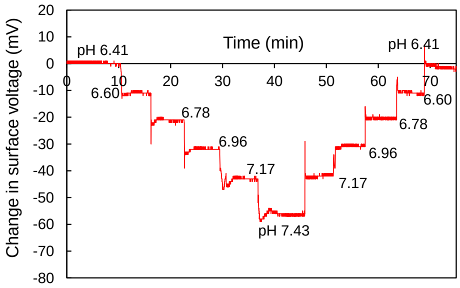


Fig. 4 Change in gate surface voltage with insulated-gate FET.

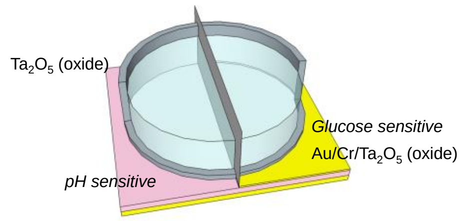


Fig. 5 Two types of extended electrodes. Ta_2O_5 oxide membrane is used for pH sensitivity, while Au film is done for glucose measurement.

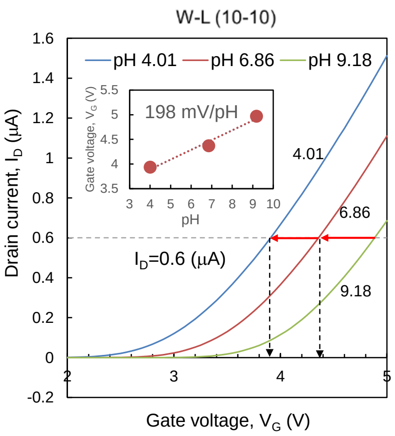


Fig. 6 V_G-I_D characteristics for pH variation with extended-gate FET.

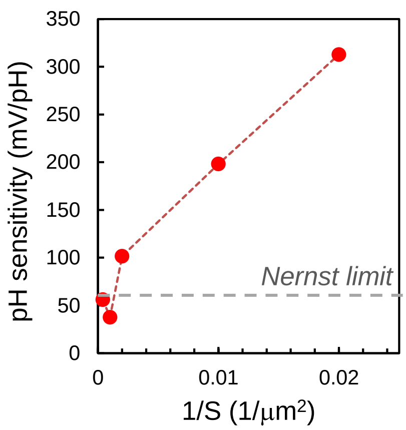


Fig. 7 pH responsibility of InGaZnO-based FET with various sizes of gate.

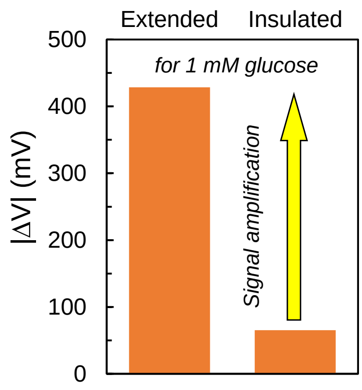
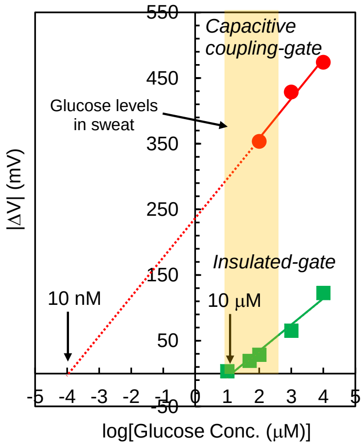


Fig. 8 Glucose response at 1 mM with InGaZnO-based biosensor.

→Fig. 9 Detection limit of glucose with capacitive coupling-gate InGaZnO-based biosensor.



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