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Characteristics of an Enzyme-based ZnO/$Zn_{0.7}Mg_{0.3}O$ Heterojunction Field-Effect Transistor as a Glucose Sensor

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1. Introduction

Zinc oxide (ZnO) has attractive advantages including low toxicity to human body, high transparency to visible light and high conformity of immobilization of organic molecules. We previously reported the formation of two-dimensional electron gas (2-DEG) at ZnO/$Zn_{0.7}Mg_{0.3}O$ heterointerface [1,2] and succeeded in applying the 2-DEG to a prototype of electrolyte-solution-gate ZnO/$Zn_{0.7}Mg_{0.3}O$ heterojunction field-effect transistor (HFET) [3]. In this presentation, application of the electrolyte-solution-gate HFET for glucose detection is discussed.

2. Experimental

Figure 1 shows schematic of a device structure and experimental setup. The ZnO/$Zn_{0.7}Mg_{0.3}O$ heterostructure was grown on an *a*-plane sapphire substrate by molecular beam epitaxy. The film was a wurtzite single crystal in the $-c$ axis (O-polar) orientation, and a 2-DEG of ~1×10^{13} cm^{-2} was induced at the ZnO/$Zn_{0.7}Mg_{0.3}O$ heterointerface by the polarization in *c* axis direction [4]. Details of the growth procedure and the electrical properties with the application to conventional type HFETs were described elsewhere [5].

On the $Zn_{0.7}Mg_{0.3}O$ surface of a HFET, ohmic contacts for drain and source electrodes were formed by indium metal to wire an external electrical power supply. These electrodes were covered by epoxy resin to protect them from electrolyte solution. The size of the exposed $Zn_{0.7}Mg_{0.3}O$ surface area was 3 × 5 mm. We modified the exposed surface with amino groups using an aminosilane agent, and then a part of the amino groups was bonded to an enzyme of glucose oxidase (GOD) by a cross-linking method using glutaraldehyde. The remained amino groups operate to detect the protons produced by a biocatalyzed reaction of the GOD molecules; β-D-glucose + O_2 $\xrightarrow{\text{GOD}}$ gluconolactone + H_2O_2 $\xleftrightarrow{H_2O}$ gluconic acid + H^+. Electrical characteristics of this enzyme-based HFET was measured in a phosphate buffered saline (PBS) solution at pH = 7.0 - 7.5 using a commercially available Ag/AgCl reference electrode.

3. Results and discussion

Drain current (I_D) vs. drain-source voltage (V_{DS}) relationship of the GOD immobilized HFET in PBS solution was found to follow conventional FET characteristics and show a clear pinch-off and current saturation region with a large on/off ratio of ~10^4. The device operation was in an *n*-type depletion mode in agreement with the presence of the 2-DEG current channel at ZnO/$Zn_{0.7}Mg_{0.3}O$ heterointerface.

Enzyme activity of the GOD molecules immobilized on the $Zn_{0.7}Mg_{0.3}O$ cap layer was analyzed by a colorimetric method using phenol, an enzyme of peroxidase (POD), and 4-aminoantipyrine reagents. Figure 2(a) shows optical transmittance spectra of a PBS solution with the GOD immobilized HFET before and after pouring 5 mg/cm^3 of β-D-glucose. A visible absorption centered at 505 nm was appeared after 1 h of the glucose input at 30°C by the chemically amplified POD associated phenol reaction with the GOD-yielded hydrogen peroxide. As shown by Fig. 2(b), even after tenth repetition of the colorimetric analysis, proportional increase of the absorbance with glucose concentration and no significant change of the absorbance intensity were observed as a direct evidence of GOD activity.

Figure 3(a) shows the change of I_D of the GOD immobilized HFET in PBS solution with an increase of the glucose concentration from 0 to 8 mg/cm^3. We found that the I_D rapidly increased corresponding to the step-like rise of glucose concentration. The time-constant of the I_D rise-up was less than 20 s. The amount of the I_D increase after 8 mg/cm^3 β-D-glucose input corresponded approximately to the pH decrease from 7.5 to 6.0 whereas the actual pH in the solution was found to remain nearly at the initial value. Therefore, it is concluded that the current response is due to the specific adsorption of the biocatalyticaly yielded protons to the amino groups on the HFET.

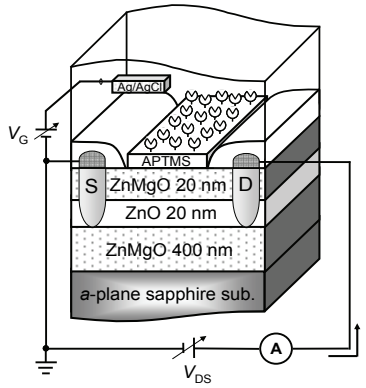

Fig. 1 Schematic diagram of an enzyme-based ZnO/$Zn_{0.7}Mg_{0.3}O$ HFET. An enzyme of GOD was immobilized using a cross-linking method on the amine-modified $Zn_{0.7}Mg_{0.3}O$ surface.

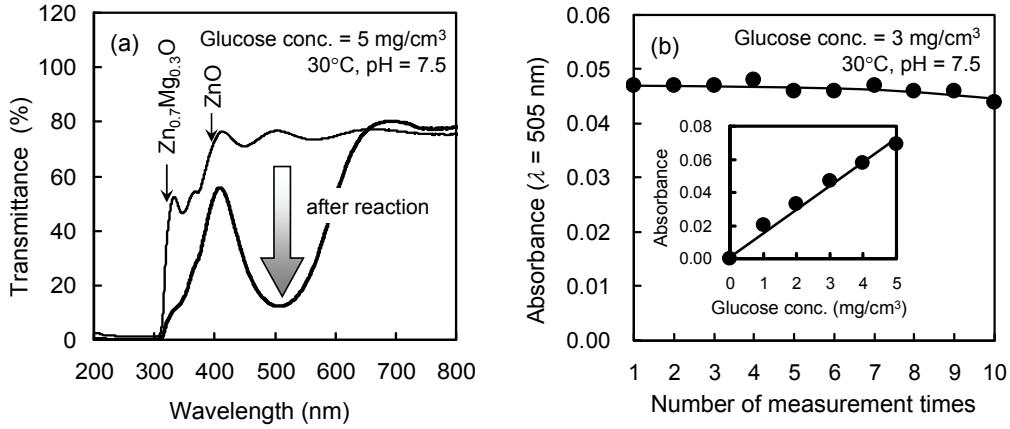

Fig. 2 Transmittance spectra of a PBS solution with a GOD immobilized ZnO/$Zn_{0.7}Mg_{0.3}O$ HFET before and after pouring 5 mg/cm^3 of β-D-glucose into the PBS solution., (a), and the absorbance reproducibility, (b). Inset shows the absorbance at 505 nm as a function of the β-D-glucose concentration.

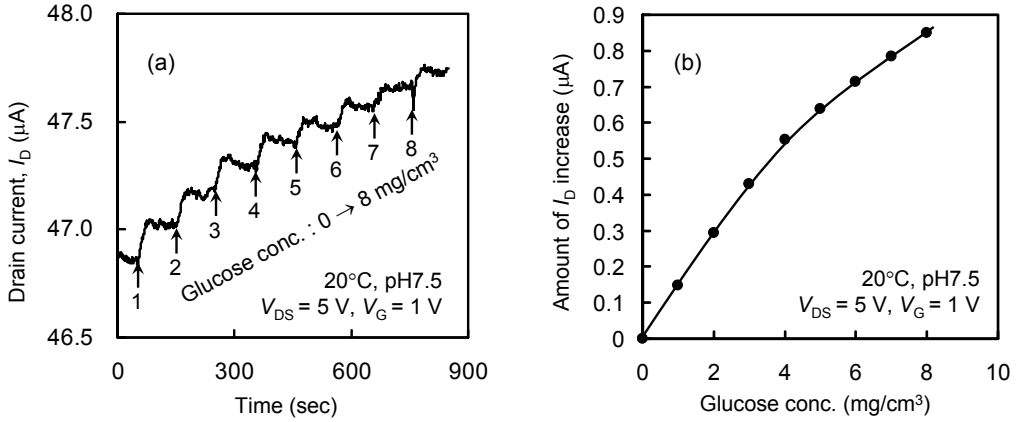

Fig. 3 Current response of the GOD immobilized ZnO/$Zn_{0.7}Mg_{0.3}O$ HFET to successive addition of a thick β-D-glucose solution to a pH 7.5 PBS solution at 20°C, (a), and the amount of I_D increase with the β-D-glucose concentration, (b).

3. Conclusions

We have succeeded in fabricating a prototype enzyme-based FET using an O-polar ZnO/$Zn_{0.7}Mg_{0.3}O$ heterostructure. An enzyme of GOD was immobilized on the amine-modified $Zn_{0.7}Mg_{0.3}O$ gate electrode using a cross-linking method. The device operated in an electrolyte solution showed sensitivity to β-D-glucose in the wide range of 0 - 8 mg/cm^3 with a fast response time less than 20 sec. The result presented here might be promising for the development of next-generation compact biosensing chips.

Acknowledgements

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