Investigation of Glucose Sensitivity of Porous Silicon

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Abstract—Accurate determination of blood glucose is essential for the diagnosis and long-term management of diabetes. Glucose and detection of its small quantities in the analyte is very important. Nowadays there is a large variety of biosensors for glucose detection working on different principles. In this study the possibility is discussed to apply porous silicon as a transducer for measuring various concentrations of glucose.

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1. INTRODUCTION

The rising demand for biosensors with high sensitivity, high reliability, fast response, electrical yield, and significant selectivity become a stimulus in carrying out research and development in semiconductor electronics. Owing to the capability of detecting trace amounts of biomolecules in real time, biosensors have found versatile applications in the field of environmental control, hazard material detection, pharmaceutics and clinical diagnostics. A common biosensor works in the following way: it interacts with a particular analyte and generates detectable signals, such as electrical, electrochemical, optical, piezoelectric and thermal responses.

While blood glucose levels in healthy individuals range up to 8 mM, people with diabetes, due to metabolic disorder in organism, may have a much larger range of approximately up to 30 mM [1]. This very large fluctuation can lead to a variety of health problems. High blood glucose levels (hyperglycemia) causes damage to the eyes, kidneys, nerves and blood vessels. A high glucose level is relatively easy to detect. Meanwhile, a low glucose level (hypoglycemia) is difficult to detect and it can lead to fainting, coma, and even death. It is necessary to frequently monitor and control one's blood glucose level to drastically reduce the incidence of these health complications in both types of diabetes.

The possibility of applications of porous silicon in chemical sensors and biosensors, also as a novel transducer material, is often discussed recently. Note that the fabrication of porous Si by means of an anodic etching process is fully compatible to the modern semiconductor technology in sensor fabrication and microelectronics. The main advantages of sensors based on porous silicon are the following:

1. By choosing the appropriate etching conditions pore sizes can be easily adjusted between some nm (microporous Si) and several tens nm (macroporous Si),
2. Since porous silicon has considerably enlarged effective surface area, which is often used as a sensing part, it's possible to miniaturize sensors down to the micron scale.
3. In contrast to ion-sensitive field-effect transistors (ISFETs), no additional passivation layer on the sensor chip is necessary [2, 3].

When using porous silicon, one has no need in additional processing the surface by biofunctionctional layer for covalent bonding of biomolecules.

2. EXPERIMENTAL

We have prepared a metal/porous silicon/metal structure for using it as a biosensor. For this purpose the surface of porous silicon was exposed to glucose of various concentrations. Current–voltage characteristics were measured for studying changes in current passing through the structure depending on the glucose concentration. We obtained the sensitivity of the sensor, which is defined as a ratio between the current passing through the structure and the surface concentration of glucose at a given voltage.

We have used <100> boron doped low resistivity p-type silicon wafers. The wafers were cut into 15x15 cm size samples. Then, porous silicon layers with 10 mm in diameter were prepared on silicon surfaces by anodic etching. Etching solution was prepared on the basis of hydrofluoric acid and ethanol in ratio HF [48%wt] : EtOH [96%], mixed 1:1 by volume. The current density during the anodization was 50 mA/cm^2, at the anodizing time 3 min. The anodization was carried out in galvanostatic regime. In order to remove contaminations from the silicon surface before the anodization, samples were first boiled in isopropyl alcohol for 5 minutes, after that washed in distilled water, and then again boiled in a toluene (also 5 minutes), with subsequent washing in distilled water. Anodizing process was performed under illumination by 1 kW power halogen lamp which was 20 cm away from the samples.

After that silver contacts were deposited onto the surface of porous silicon by thermal deposition method. The contacts had crested shapes as shown in Fig.1. The distance between the two contacts was 2 mm. After measurements porous silicon structures were easily restored by heating to 350^{O}C in vacuum during 10 minutes. But new calibration of the device is required.

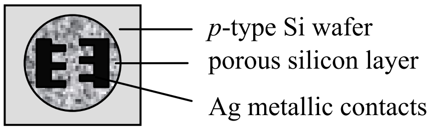

Fig. 1. Porous silicon with metallic Ag-contacts.

3. RESULTS AND DISCUSSION

The samples were dipped into glucose solution with various concentrations. We used glucose commonly employed in medicine. It was diluted in distilled water at different concentrations ranging from 10 to 1000 μg/l. The structures were incubated in 2 ml glucose solutions for 20 min. According to [3], removal of non-adsorbed glucose molecules was accomplished by immersing the samples in 2ml of distilled water for 5 min. This process was repeated twice. So, only immobilized glucose molecules remained on porous silicon surface. The surface of porous silicon was investigated with use of scanning electron microscope (SEM) VEGA TS 5130MM. Fig.2 shows a typical SEM image.

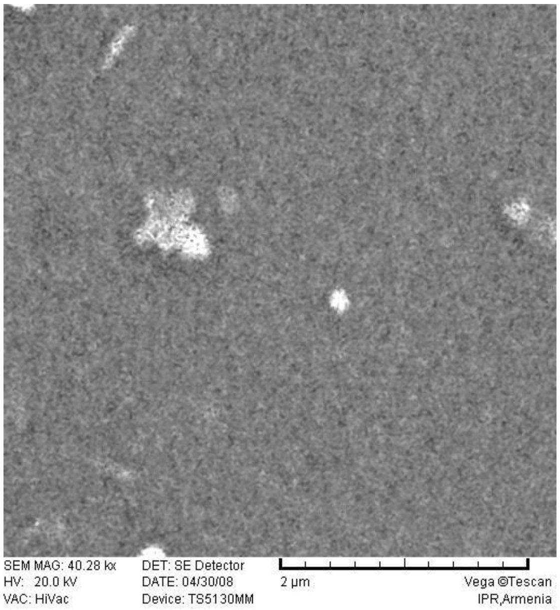

Fig. 2. SEM image of the surface of porous silicon with immobilized glucose molecule.

Figure 2 demonstrates clearly immobilized glucose on the porous silicon surface, and we can assume that appreciable porosity of silicon provides immobilization of glucose, so in case of using silicon as a sensitive element (part) no additional special biofunctional layer is needed.

As known, porous silicon has a large bio-activity [4] because of its specific surface which provides numerous available bonding sites, as a result of the electrochemical formation process in hydrofluoric acid based solutions. Also note that silicon is biocompatible with organism [2, 3].

Current–voltage measurements were performed with use of pressure contacts by the instrumentality of automatized measurement scheme connected to the computer. Figure 3 shows the current–voltage characteristics of the structure after exposure of various glucose concentrations on porous silicon.

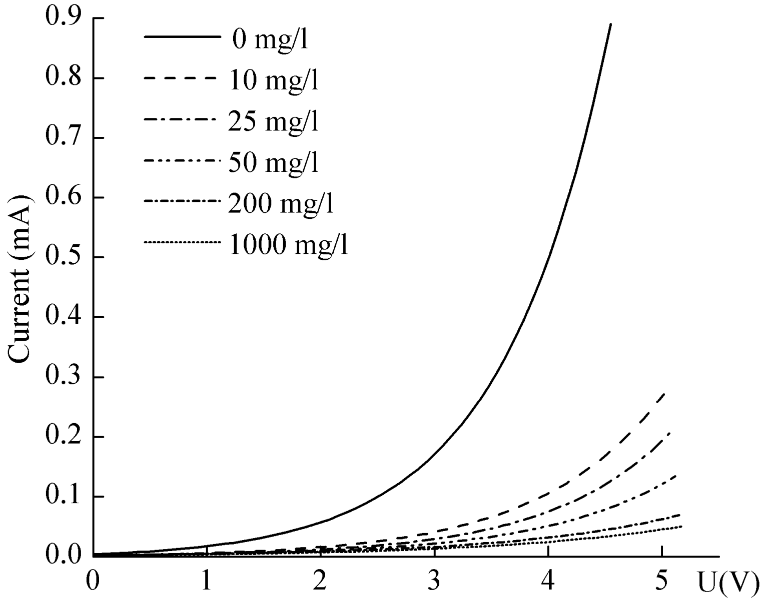

Fig.3 Voltage dependence of current at different glucose concentrations.

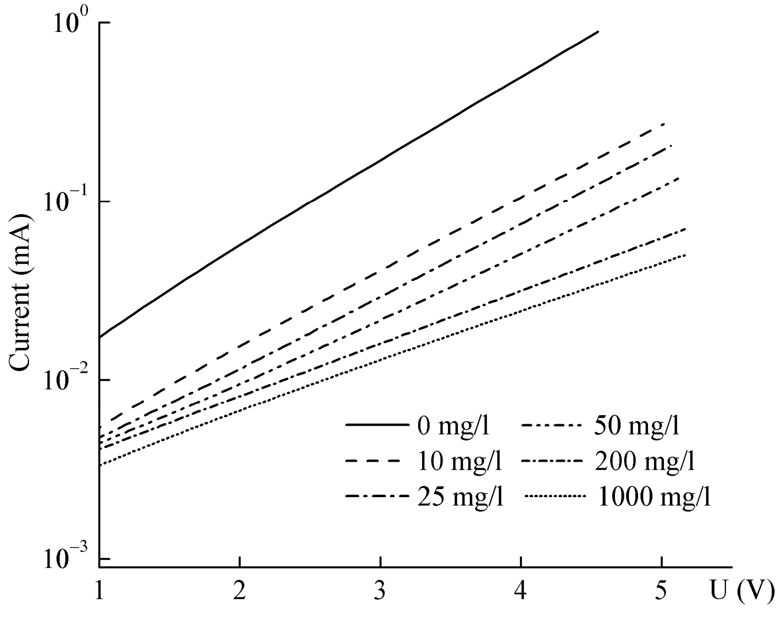

Fig.4 Log-plots of current versus voltage at different glucose concentrations.

As seen in Fig.3, the current–voltage characteristics of such structure are asymmetric and have rectifying behavior which observed in all tested glucose concentrations. It should be noted that with the increase in the concentration of immobilized glucose the current–voltage characteristics become closer to the voltage axis, i.e., the glucose concentration growth leads to reduction of the current at a given value of the voltage. On the basis of these data, the curves showing relationships between the current and glucose concentration were plotted at specified values of voltage. Results are shown in Fig.5.

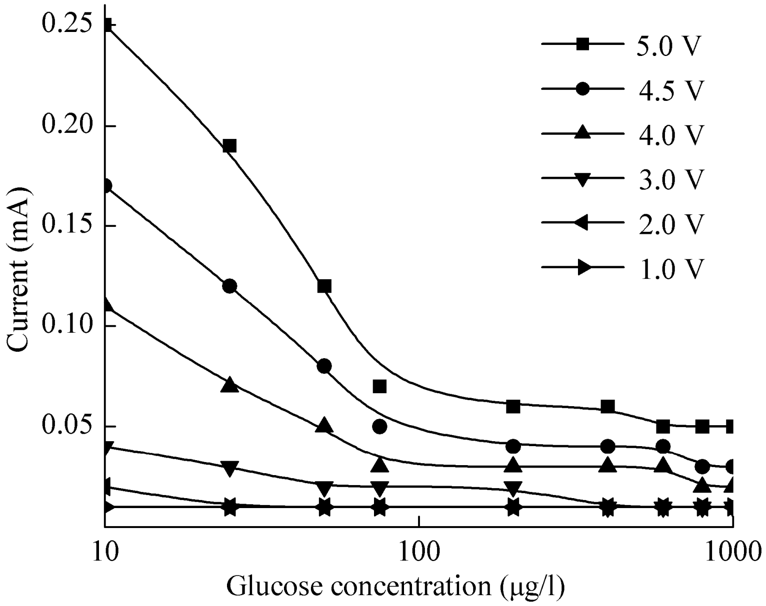

Fig. 5. Current versus glucose concentration at different applied voltages

As seen in Figure 5, dependences change slightly after some concentration (100 μg/l) and curves are almost parallel at different voltages. But the current changes rather sharply at a small glucose concentrations. From logarithmic representation of current–voltage characteristics (Fig.4), we obtained sensitivity curves of the biosensor at a specified values of voltage applied to the structure (Fig.6).

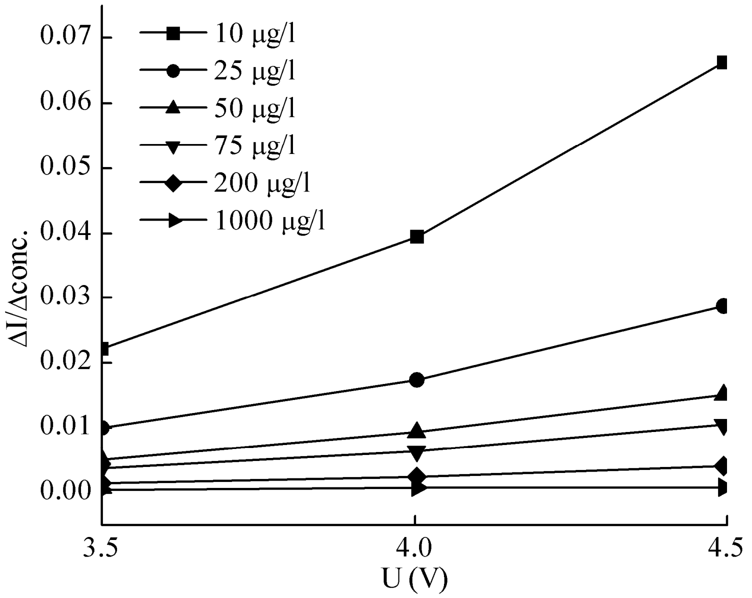

Fig. 6. Sensitivity curves of the biosensor at specified values of voltage

Hence, we can conclude that porous silicon-based biosensor structure proposed by us is especially sensitive at small glucose concentrations. It is very important for medical and clinical applications, when it is necessary to measure small quantities of glucose.

In addition, the reduction of porous silicon conductance with the increase in the concentration of glucose is a result of appearing of immobilized glucose molecules on the surface rather than dissolution of glucose in distilled water, since water, on the contrary, increases the conductivity of porous silicon.

Obtained dependences are similar to the recently reported results [4]. However, it should be noted that measured values of current are in the range of 0.01–1 mA. This is worthy of special attention because in this case there is no necessity to use special equipment for detecting very small values of current; this may be done with simple and cheap instruments.

4. CONCLUSION

Porous silicon-based devices were fabricated and their current–voltage characteristics under influence of glucose with different concentrations were measured. Glucose molecules were immobilized on porous silicon surface from solutions of glucose in distilled water. It was shown that increasing glucose concentration leads to reduction of current values at given voltages.

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