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Polycrystalline silicon ion sensitive field effect transistors

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We report the operation of polycrystalline silicon ion sensitive field effect transistors. These devices can be fabricated on inexpensive disposable substrates such as glass or plastics and are, therefore, promising candidates for low cost single-use intelligent multisensors. In this work we have developed an extended gate structure with a Si_3N_4 sensing layer. Nearly ideal pH sensitivity (54 mV/pH) and stable operation have been achieved. Temperature effects have been characterized. A penicillin sensor has been fabricated by functionalizing the sensing area with penicillinase. The sensitivity to penicillin G is about 10 mV/mM, in solutions with concentration lower than the saturation value, which is about 7 mM. © 2005 American Institute of Physics. [DOI: 10.1063/1.1854192]

The use of ion-sensitive field effect transistors (ISFETs) to measure pH and to sense a variety of ions is well known.¹ In these devices the gate insulator is in direct contact with the electrolyte solution. The high surface buffer capacity of the insulator¹ leads to an almost constant proton density at the surface, independently of the electrolyte's pH. Hence a pH-dependent potential drop across the double layer arises, in order to equalize the free energy difference for protons at the interface and in the bulk of the solution, resulting in modulation of the channel conductance.

By coating the gate insulator with enzymes as the selecting agent, highly selective sensors have been developed.² Such enzyme-modified ISFETs (EnFETs) can in principle be constructed with any enzyme that upon reaction with the analyte induces a local change of pH. ISFET biosensors are used in medical diagnostics, environmental monitoring, and food quality control. One distinctive advantage of ISFETs is their suitability for miniaturization, since the signal to noise ratio is independent of the ISFET area. Therefore large multisensor arrays capable of detecting different species and simultaneously measuring relevant parameters (temperature, pH, etc.) are possible. In all these applications cost and single-use, that is disposability, are paramount considerations.

We report here the use of extended gate polycrystalline silicon thin film transistors (poly-Si TFTs) as ISFETs and penicillin sensitive EnFETs. The poly-Si TFT technology is well established in the area of flat panel displays. In addition to the pixel switching array, where typically some 10^6 poly-Si TFTs are employed, complex integrated circuits, such as logic drivers and SRAMs can be fabricated with these devices. Furthermore, these circuits can be made on inexpensive disposable substrates such as glass or plastic.³ Poly-Si TFTs are, therefore, promising candidates for low cost single-use intelligent multisensors. In view of the fact that the device is used in an aqueous environment, an added advantage of this technology over conventional bulk CMOS

is the electrical isolation between the individual transistors.

The inset in Fig. 1 shows the schematic structure of the extended gate ISFET. The devices were fabricated on glass substrates by laser recrystallization.³ Si_3N_4 was deposited by plasma enhanced chemical vapor deposition (PECVD) on top of the tantalum extended gate, to serve as the pH sensitive layer. To provide electrical and chemical isolation, the entire device area, except the sensitive Si_3N_4 pad, was passivated with a Si_3N_4 (100 nm)/ SiO_2 (500 nm) layer. p-channel TFTs with different gate sizes have been fabricated and tested. The channel length of the TFTs is 6 μm , while the channel widths range between 10 and 100 μm . The sensing pad area is 1 mm $\times$ 1 mm.

For the penicillin EnFET, the enzyme penicillinase was adsorptively immobilized on the surface of the Si_3N_4 pad. For the immobilization, the enzyme solution was prepared by dissolving 2000 units of enzyme penicillinase in 1 ml of 50 mM phosphate buffer with pH 7.5. 10 μl of enzyme solution was pipetted onto the chip, covering one Si_3N_4 pad, and incubated for 1 h at room temperature. The sample was then rinsed and dried at room temperature. The penicillin G solu-

FIG. 1. I_D - V_{GS} curve for a p-type ISFET with transistor size $W/L = 20 \mu\text{m}/6 \mu\text{m}$ measured with $V_{DS} = 0.1 \text{ V}$ in phosphate buffer with different pH values (from left to right: pH=4.0, 7.0 and 10.0). Inset: Schematic structure of the extended gate ISFET.

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FIG. 2. (a) I_D - V_{GS} curve of a p -type ISFET with transistor size $W/L = 100 \mu\text{m}/6 \mu\text{m}$, measured with $V_{DS} = 0.1$ V in a 5 mM phosphate buffer pH 7.6 at different temperatures. The temperatures corresponding to the curves from left to right are 28.0, 30.7, 34.0, 38.5, 44.5, 48.5, 53.5, and 58.0 $^{\circ}$ C, respectively; (b) temperature dependence of the gate voltage corresponding to the different values of I_D .

tions were prepared in different concentrations, using a 5 mM phosphate buffer with pH 7.0. After each concentration step, the sensor was rinsed with the working buffer. Phosphate buffer, penicillinase, penicillin G were purchased from Sigma-Aldich.

The I_D - V_{GS} characteristics of the ISFETs and penicillin EnFET were measured using a HP4378 parameter analyzer. An Ag/AgCl reference electrode was used. During the measurements, V_{DS} was kept constant at 0.1 V and V_{GS} was swept from negative voltage to positive voltage and back. It was found that in order to avoid transient effects due to the sensor response, the sweeping speed had to be lower than 0.1 V/s. So the sensor response time to V_{GS} change is of the order of 1 s, which can be attributed to the mechanism at the Si_3N_4 /electrolyte interface, since the TFT response is much faster.

The I - V curves are very stable and repeatable and show a rigid shift with the change of pH value. The voltage shift is proportional to the change in pH. Typical characteristics are shown in Fig. 1 for a TFT with $W/L = 20 \mu\text{m}/6 \mu\text{m}$. The pH sensitivity is 54 mV/pH at 300 K, close to the ideal value of 59 mV/pH.⁴ In some cases a lower pH sensitivity, between 47 and 54 mV/pH was observed. It is expected from the site-binding theory⁵ that the state of the surface of the Si_3N_4 is critical for the pH sensing properties. Impurities, such as oxygen or other ions, which might exist at the Si_3N_4 surface⁶ can result in a nonideal sensitivity. Further investigation of these aspects is needed.

The temperature dependence of the ISFET characteristics has been measured using a 5 mM phosphate buffer with pH 7.6 and is shown in Fig. 2(a) for a TFT with W/L

FIG. 3. Gate voltage shift ΔV_{GS} taken from the I_D - V_{GS} curves of a p -type EnFET with transistor size $W/L = 100 \mu\text{m}/6 \mu\text{m}$ measured in a 5 mM phosphate buffer pH 7.0 vs penicillin G concentration.

$= 100 \mu\text{m}/6 \mu\text{m}$. Both threshold voltage and shape of the I - V curves change with temperature. Figure 3 shows the voltage shift versus temperature for three values of I_D . The behavior is linear and the temperature coefficients for the three curves are slightly different. The temperature coefficient is higher than the one observed for single crystal ISFETs.⁷ There are three main possible contributions to the observed temperature effect: (1) temperature-dependent potential change at the Ag/AgCl reference electrode; (2) temperature-dependent potential drop at the Si_3N_4 /electrolyte interface; (3) temperature-dependent poly-Si TFT characteristics. The first two contributions can only induce a parallel shift of the I - V curves, while the third one can also change the shape of the I - V curve itself.

The temperature dependence of the potential at Ag/AgCl reference electrodes has been reported to be on the order of 0.14 mV/K.⁷ On the other hand, the potential drop φ_0 at the interface between Si_3N_4 and the electrolyte is, according to site-binding theory,^{1,4,5}

$$\varphi_0 = 2.3 \frac{kT}{q} \frac{\beta}{\beta + 1} (\text{pH}_{\text{pzc}} - \text{pH}),$$

where pH_{pzc} is the pH value at the point of zero charge in the interface, k is the Boltzmann constant, q is the electronic charge, T is the temperature of the system, and β is a parameter, which can be expressed in terms of the acidic and basic equilibrium constants of the related surface reactions. Using the value of $\text{pH}_{\text{pzc}} = 5.2$ for Si_3N_4 ,⁵ the temperature coefficient becomes 0.48 mV/K at pH 7.6. The values for the temperature coefficients for the first two possible contributions described above are much smaller than the experimentally observed one. This value is consistent with the temperature dependence of poly-Si TFT I - V characteristics. This dependence is controlled by the gap density of states, which is fairly reproducible in a stable process. Therefore a reference TFT can be used as a temperature sensor and to provide the input for temperature compensation.

For the penicillin EnFET, penicillinase was immobilized on the Si_3N_4 gate dielectric. The functionalized ISFET detects the variation in H^+ concentration, resulting from the catalyzed hydrolysis of penicillin by penicillinase, which is dependent on the penicillin concentration in the solution.^{2,8} Figure 3 shows the voltage shift, measured in 5 mM pH 7.0 phosphate buffer versus penicillin G concentrations. The I - V curves shift to negative voltages for increasing penicillin

concentration. The shift is linear with concentration until saturation is reached at about 7 mM. The sensitivity is 10 mV/mM.

In conclusion, we have fabricated ISFETs on glass substrates, by using low-temperature Poly-Si TFT technology. An extended gate structure has been developed, using PECVD Si_3N_4 as the pH sensitive area. Stable operation and nearly ideal pH characteristics have been obtained. A penicillin G sensor has been fabricated by functionalizing the Si_3N_4 with penicillinase. The extended gate structure facilitates the electrical and chemical isolation of the sensor, hence increasing its stability and durability. This technology, in view of the advanced level of development for mass production of polysilicon TFTs and the use of cheap substrates such as glass or plastics, shows great promise for application in single-use multi-analyte biosensors.

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