



Organic field-effect transistor with extended indium tin oxide gate structure for selective pH sensing

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ABSTRACT

In this study, organic field-effect transistors (OFETs) with extended gate structure were fabricated for selective pH sensing applications. Indium tin oxide (ITO) was used as extended gate electrode as well as an active layer for H⁺ sensing. The threshold voltage of the fabricated ion-selective OFET was varied by the changes in the electrochemical potential at the ITO electrode surface upon its exposure to buffer solutions with variable pH values. The sensor showed excellent linearity and a high sensitivity of 57–59 mV/pH in the pH range of 2–12. The selectivity of the ITO sensing layer to H⁺ ions was also investigated by measuring the interfering effect of Ca²⁺ and K⁺ ions in the buffer pH solutions. The results showed that the Ca²⁺ and K⁺ ions weakly interfere with the selective pH sensing of the ITO-extended gate OFET sensor device.

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1. Introduction

In bio-chemical processes, it is important to know the basic conditions of electrolyte or physiological solution. In this respect, the pH sensor for detection H⁺ concentration was developed long time ago. For several decades, it has been still being investigated for the purpose of improving its performance which is characterized by parameters such as the sensitivity, selectivity, stability, drift–hysteresis effect, fast response, lifetime, and cost of fabrication.

The popular structures for pH sensors are based on selective glass membrane electrode and Si ion-sensitive field-effect transistor (ISFET) structures [1,2]. The selective glass membrane pH electrode has been the most frequently used pH indicator for many years, due to its high selectivity and dynamic pH range. However, it has the disadvantages of high resistance, brittleness, and difficulty to integrate into a miniaturized system [2]. Since the Si ISFET

for chemical sensing was first introduced by Bergveld in 1970 [1], it has been extensively researched and commercialized [3]. The first Si ISFET was based on the use of a silicon dioxide (SiO₂) gate dielectric in the MOSFET (metal-oxide-semiconductor FET) structure as the H⁺ sensing material. Since then, many other dielectric materials have been investigated for the purpose of replacing SiO₂ as the sensing layer for other ions, as well as H⁺, such as Al₂O₃, Si₃N₄, Ta₂O₅, ZrO₂, HfO₂, Y₂O₃, and Gd₂O₃ [4–8].

However, there were still drawbacks in these structures, because in the Si ISFET structure there is poor isolation between the gate dielectrics and the chemical solutions, due to the direct exposure of the sensing area to the solutions. This problem causes difficulties in its fabrication and has limited its chemical and biological applications, because of the interaction of the dielectric layer with the solution, resulting in the instability of the sensor. These disadvantages could be overcome by using the extended-gate ISFET structure that was firstly introduced by Spiegel in 1983 [9]. In this structure, the sensing area is on the extended-gate side away from the active area of the ISFET structure. Therefore, it is easy to deposit or modify the sensing materials by various methods that have no effects on the

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performances of the ISFET device. A pH sensor that combined the commercial MOSFET and an extended-gate concept using V_2O_5 xerogel, tin oxide (SnO_2) and indium tin oxide (ITO) as the pH sensing material was investigated by various authors [10–12]. The pH probe coated with V_2O_5 , SnO_2 , and ITO as the pH sensing materials electrically connects with the extended gate electrode of the commercial MOSFET devices. The reference electrode used for the measurement of the FET characteristics is used as a floating electrode, while both the reference electrode and pH probe are immersed in the electrolyte. However, the selectivity of these sensing layers to other ions including K^+ and Ca^{2+} was not measured.

Despite the various advantages of the Si ISFET device, it also has some limitations, due to the rigidity and opacity of the Si substrate and difficulty of achieving monolithic integration with microfluidic components for lab-on-a-chip (LOC) or micro-total analysis systems (μ -TAS) for clinical use. For the facile integration of ISFET devices into the new concepts of LOC or μ -TAS based on transparent polymeric or glass substrates having the capability of electrochemical and optical detection, new ISFET devices based on thin film transistor technologies from the display industry look promising. Among the various ISFETs based on non-single crystalline Si channels [13–15], the organic FET (OFET) is a good candidate, because the OFET has shown considerable potential in many electronics applications, due to its light weight, low cost, mechanical flexibility, optical transparency, compatibility for large area applications, and simple fabrication compared to conventional MOSFET devices based on silicon [16,17]. Bio-chemical sensors based on OTFTs have been developed by functionalizing an organic semiconductor sensing layer to specific target species. Some groups have also developed organic ISFETs for H^+ sensing [18,19]. However, the application of organic ISFETs is still limited, because of their limited dynamic pH range from 4–10 [18–20], poor linear sensitivity behavior [18–20], slow response [19], and poor stability [18]. These issues are related to the inherent degradation of the organic semiconductor layers which are sensitive to moisture and oxygen. However, in the above-mentioned cases, the organic dielectric or semiconductor layers are directly exposed to

the electrolyte. Also, organic ISFETs with a top gate structure have difficulties in terms of the fabrication of the H^+ sensing gate dielectric on top of the organic semiconductor layer, because of the damage incurred during the formation of the gate dielectric. The damage to the organic semiconductor can be prevented by using organic gate dielectrics layers, such as parylene or polyvinyl alcohol (PVA) layers. However, choosing oxide gate materials for H^+ sensing is challenging in this case, due to the large difference in the thermal expansion coefficients of inorganic and organic materials. Furthermore, the ions can be transported through the gate insulator, which gives rise to a leakage current, due to the formation of cracks in the sensing layer under the influence of the large electric field in the solution during the measurement. These factors will affect the stability of the sensor performance.

In our work, a bottom-gate OTFT structure with ITO-extended gate sensing electrode was successfully fabricated. The side-effects caused by the fabrication process were minimized to improve the stability of the organic ISFET for pH sensing. This structure could be used to develop a stable pH sensor with good linearity over a wide dynamic pH range of 2–12, high sensitivity of 57–59 mV/pH, and fast response. The ITO-extended gate electrode in the organic ISFET could be used for sensing the pH selectively in K^+ and Ca^{2+} containing solutions.

2. Experiments

Fig. 1 shows the cross-section view (a) and top view (b) of the pH sensor based on the ITO-extended gate OFET. The ITO glass substrates were purchased from Fine Chemicals Co., Ltd., (Korea). The resistivity of ITO is below 20 ohm/sq, its thickness is 80 nm, and its transmittance is 87%. The ITO-extended gate OFET with a channel length of 40 μ m was fabricated on an ITO-coated glass substrate. The ITO layer on the ITO-coated glass substrate was selectively etched by an etchant solution consisting of 24 wt.% HCl followed by a photo lithography process for patterning to form a gate electrode for the OFET as well as H^+ sensing layer. Then, poly(4-vinylphenol) (PVP) was deposited by the spin coating pro-

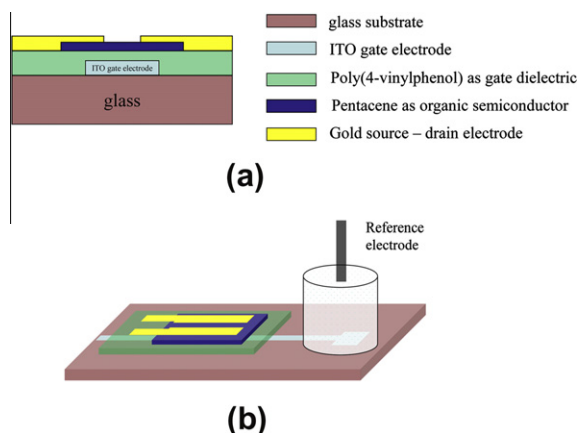


Fig. 1. Schematic of cross-section and top view of ITO-extended gate OFET device.

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