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A review of the application of thin film transistor in ph sensors

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Abstract

This paper has reviewed the recent research progress on the application of thin-film transistors in pH sensors, specifically on aspects related to the structures and materials, including active channels and the top gate dielectrics. Results indicate that the pH sensitivity can be promoted by the utilization of specific structures. Moreover, employing proper top gate dielectrics and active channels can promote the capacitance ratio of the top-gate dielectric to the bottom-gate dielectric, such as the a-IGZO as the active channel and Ta_2O_5 as the top gate dielectric.

Keywords: ph sensors, thin film transistor, structure, material

Introduction:

Thin-film transistors (TFTs) have been inclusively employed in different sensors. In the near future, the application of sensors will promote their sundry functions in different areas to make important contributions to mankind and benefit human a lot as well. Because of this, the requirement for sensors is increasing. Particularly, biosensors are usually employed to detect different biological indicators such as blood acidity and alkalinity, glucose concentration and so on, in order to make a diagnosis of diseases and supervise people's health [1]. The emergence of other transistors are due to the first ion sensitive field effect transistor (ISFET). Almost four decades, ISFET sensor has been widely investigated and has been changing, employed to be a transducer, with its various advantages of fast response to ions change, low cost to manufacture, and compatibility with the advanced CMOS fabricating technologies etc.. The ISFET is a kind of potentiometric sensor initially designed for the detection of hydrogen ions, which can detect the change of the interface surface potential φ_0 between dielectric and electrolyte [2]. However, the ISFET sensor consists of the detector and the transducer in one device, when the detector testing, the chemical solution will suffer damage and the chemical solution also damages the transducer. What's more, other weakness of the ISFET sensors is its traditional single gate structure cannot break through the Nernst limit of 59mV/pH at room temperature [1]. To overcome these major problems, kinds of structures have been proposed and various materials are used by experts. In our review article, we first discuss the structures of the thin-film transistor in pH application in recent years. In the second section, we discuss material that is used to promote the sensitivity of pH sensors.

Structure of pH sensors：

Ion-sensitive field-effect transistor pH sensors: P. Bergveld firstly reported the ISFET device in 1970. The device is combined the working principles of MOS transistors and a glass electrode, which can take the advantage to measure the ion activities in electrochemical and other specific environments. [1]. The vital difference compared with traditional measuring ways is that with the glass electrode mentioned above, which can be regarded as an ISFET, and the reference electrode is avoided to use. Besides, most importantly, the channel resistance of this structure is low ohmic, which can be compared with the measurement of light-sensitive and pressure-sensitive resistors and so on. However, living ISFETs has some deficiencies. Due to the low isolation of ISFETS, chemical stability of them is fragile and the disposable sensor is insufficient, which makes sensitivity change easily, and particularly combined structure (of detector and transducer). Moreover, the Nernst limit confines the essential pH sensitivity of ISFET, which describes a theoretical maximum 59 mV/pH at 298k [2, 3].

Heterostructure pH sensors: In recent decades, the gallium nitride (GaN) as the representative of third-generation semiconductor has been extensively applied in electronic devices, optoeletronic devices as well as the ion-sensitive heterostructure transistors based pH sensors. But the sensitivity of GaN-based semiconductors are still required to improve. It is stated that the indium nitride (InN)/GaN-based heterostructured pH sensors whose surface was passivated by photoelectrochemical (PEC) method showed a pH sensitivity of 52.04mV/pH with a linear pH response [1]. The AlGaN/GaN-based heterostructure pH sensors with the deposition of a passivation layer on the surface of AlGaN obtained a higher pH sensitivity of 55.6 mV/pH [2]. Besides, AlGaN/GaN-based heterostructure pH sensor owing multi-segment sensing regions also has been fabricated and characterized. A much higher pH sensitivity of 1.35 mA/pH gained in the AlGaN/GaN-based pH sensors with multi-segment sensing is higher than conventional AlGaN/GaN-based pH sensors with single sensing area. Roughly, the sensitivity of the multi-sensing sensor can be increased by widening the sensing area locating in the gate region which, in contrast, increases the avoidable noise so that decreases the pH sensitivity as well as the chemical stability of the sensor [3].

Electrolyte-Gated pH sensors: TFT-based pH sensors with a top-gate dielectric have been widely studied, taking the advantages of low cost, ease fabrication, and are compatible with flexible devices and can be integrated possibly on chips. However, the fundamental pH sensitivity of ISFET is confined by the Nernst limit of 59 mV/pH at 298k. Lately, as a pH sensor, the electrolyte-gated thin film transistors (EGTFTs) have been studied. Without any ion-sensitive dielectric layers, the structure of EGTFT is very simple. The chemical solution is directly contacted with the active layer. However, by taking a specific passivation treatment on active layer, EGTFT pH sensor with In_2O_3 active layer obtains the high sensitivity as high as 75mV/pH, which is beyond the Nernst limit [1]. Using the active layer, amorphous indium-gallium-zinc oxide (a-IGZO), as the pH active layer in ISFETs could avoid using a high-κ dielectric and cut down a few of fabrication steps [2]. The thin-film transistor (TFT) with a dual-gate IGZO investigated obtained a pH sensitivity of ~ 160 mV/pH, which was over Nernst limit [2]. Additionally, a group presented using a chemical layer the branched polyethylenimine (BPEI) coated a-IGZO based EGTFT which obtained a high pH sensitivity of ~ 160 mV/pH exceeding the Nernst limit [3]. In general, researchers utilize the electrical double layer (EDL) formed at the electrolyte/active layer interface provided the function of the top gate oxide capacitance to obtain notable capacitive coupling in the functioning of the dual gate TFT based ISFETs [4]. Therefore, they obtain a high sensitivity above the Nernst limit. However, electrolyte-gated pH sensors will face a serious problem that their active channels are directly contact with the buffer solution, and thus, will lead to a limit to the durability of pH sensors.

Double-Gated pH sensors: To overcome above shortcomings, a top dielectric is added upon the active channel to protect the channel directly contact with chemical solution. To enhance pH sensitivity, a high capacitance ratio of the top-gate dielectric to the bottom-gate dielectric must be valued. That is also why double-gated(DG) pH sensors always gain a high sensitivity. Employing the high capacitance ratio between the top-gate insulator and the bottom-gate insulator, the sensitivity of ZnO ISFET is enhanced from 22 mV/pH to more than two orders of magnitude up of 2.25 V/pH [1]. With an enhanced chemical stability, the ultra-thin-body DG ISFET exhibits a conspicuous sensitivity of 425.89 mV/pH [2]. DG IGZO TFTs are fabricated and titanium dioxide (TiO_2) is deposited by atomic layer deposition (ALD) at low temperature (< 180°C) and a sensitivity of 76 mV/pH is gained, which is over the Nernst limit at 298k [3]. DG IGZO TFTs are fabricated using tantalic oxide (Ta2O5) deposited via e-beam evaporation followed by annealing. The achieved pH sensitivity of the DG based ISFET was 402 mV/pH, which outclassed the Nernst limit (59 mV/pH) [4]. The pH sensitivity of the a-IGZO ISFET fabricated at the 40 nm hafnium oxide (HfO_2) sensing layer in the DG mode obtains a pH sensitivity of 937 mV/pH, which is far more than the Nernst limit [5]. However, because the ISFET sensor consists of the two most significant part, detector and transducer, any damage to the detector caused by the chemical solution being tested also damages the transducer. Nevertheless, to overcome this disadvantage, the structure of the extended gate FET (EGFET) was invented. With using the EGFET, TFT can easily isolate the FET from the chemical solution. The sensitive layer is exposed to chemical solution or biological environment in other structures, but it is isolated from the electrically connected FET device in the EGFET. Generally, the EGFET includes two different parts: one is the sensing membrane electrode, and the other one is the commercial MOSFETs. This structure is very suitable for application as disposable biosensors, because the specific structure allows MOSFET to be reusable when the sensing part needs to replace. So far, a-IGZO based TiO_2 extended gate field-effect transistors (EGFETs) in the dual gate operation mode obtains a high pH sensitivity of 129.1 mV/pH over the Nernst limit [6]. With a dual gate operation mode, a-IGZO SEGISFET achieved a higher pH sensitivity of 649.04 mV/pH [7]. With Ta_2O_5 sensing layer having high permittivity, the DG EGFET exhibits a high pH sensitivity of 478.0 mV/pH [8]. By and large, the structure of EGTFT is an appropriate choice when we are considering cheap and disposable biosensors.

Triple-Gated pH sensors: Apart from the dual-gated TFTs, another structure under the spotlight is the triple-gated TFT which has a wonderful sensitivity [9-12]. A Triple gate ISFET is a transducer device which has three gates including a top gate on the channel, a bottom gate under the channel, and a triple gate on the same plane of the channel. As a result, the pH sensitivity of triple gate (TG) mode using the triple gate measured is much higher than conventional single-gate (SG) mode measurement using the top-gate, or dual-gate (DG) mode measurement using the bottom-gate. The measured pH sensitivity was 57.75 ± 0.77 mV/pH in the SG mode, 467.08 ± 9.92 mV/pH in the DG mode [13-17]. While, the TG mode gave a maximum sensitivity of 1283.56 ± 45.54 mV/pH for a sensing membrane having a theoretical Nernst limit(59.15 mV/pH at 25 °C) [18-21]. Even for a small in-plane-gate area, the fabricated In-plane-gate pH sensor exhibited a good linearity about 99%, and with reducing in the IPG area, the amplification of the sensitivity could be promoted. Notably, a high sensitivity of 387.58 mV/pH has been achieved outclassing the Nernst limit, at the lowest IPG area. Moreover, a low-cost disposable biosensor combined a low-cost extended gate (EG) detector with an ion-sensitive membrane to the triple gate ISFET transducer [22-25]. In a word, the EGFET with the structure that isolates the FET from the chemical environment, triple-gate pH sensors have all-round reinforcements in sensitivity and other aspects.

Material of pH sensors：

Active Channels： For the ISFET, the active channel is an essential part. In this respect, to replace the traditional amorphous silicon (a-Si), zinc oxide (ZnO)-based TFTs have been widely investigated as the potential alternatives, which can give a reasonable switching capability even when fabricated at low temperatures below 300 °C [26-28]. The use of single crystal ZnO on the TFTS was reported in 1968 [29]. But it is the one believed difficult to form the amorphous or single crystalline ZnO thin films, and generally a polycrystalline structure is obtained, leading to form the grain boundary defects [30]. Due to the defects existing, non-uniform TFT will perform at different areas across the flat panel display. Another problem is that the fabrication of ZnO thin films is not easy, for the chemical durability of pure ZnO against chemical environment is low [2]. Many papers make use of the a-IGZO as active channel. In 2004, a great invention was reported that Nomura et al. utilized a new kind of amorphous oxide semiconductors based on IGZO deposited at room temperature, showing the high-performance transistors ($\mu \approx 8.3$ cm^2/Vs) [31]. Regardless of the amorphous state, the electronic orbital structure of the material will make a high mobility to IGZO based TFT. The conduction band in IGZO is mainly formed by the overlap of the In 5s orbitals, which exhibit isotropic properties. In comparison with Si that undergoes significant reduction in mobility from 1000 (single crystal) to 1 (amorphous) cm^2/Vs. Apart from this, to give the optimal performance of IGZO, incorporating Ga ions act as a carrier suppressor and network stabilizer in the conductive IZO host [32].

Top gate dielectric: For thin-film transistors used in ISFET, the top-gate dielectric protects the active channel directly from contacting the buffer solution and realizes a high capacitance ratio with the bottom-gate dielectric. Hereinto, the high dielectric constant material is significant in oxide semiconductor TFTs. There are various kinds of high-κ dielectric materials recently studied as sensitive layer in ISFET based TFTs. However, these materials can easily react Si substrates so that a silicate layer will be formed at the interface of the oxide film/Si substrate, thus reducing pH sensitivity performance of the sensors [15-19]. It is universally acknowledged that the interface of dielectric layer affect the sensor's performance significantly which can protect the sensor from the damage of chemical solution. Consequently, this calls for academia to find alternative or new materials and better processes to decrease interfacial defects. Nowadays, a growing number of papers demonstrate that rare earth (RE) oxide films are possible substitutes for traditional SiO_2 as the gate dielectric in advanced CMOS devices and in biochemical pH sensors because of their advantages, such as large values of κ, good thermal stability and so on [2-4]. Among them, cerium oxide (CeO_2) thin film performs remarkably, having the advantage of strong adhesion, good refractive index, stable redox, high mechanical strength, excellent thermal stability, for applications in humidity sensors, fuel cells and gas sensors and so on[5, 6]. In order to avoid the moisture absorption, several scholars have studied various methods, such as yttrium oxide (Y_2O_3) incorporating, titanium oxide (TiO_x) doping, or Ti adding into rare-earth oxide films [7-9]. Survey data showed that, above metioned materials show a high sensitivity. It is reported that amorphous Ta_2O_5, in addition to possess a high dielectric constant of ~ 25, shows good pH sensitivity performance [10-12]. With engineered gate oxides, a-IGZO TFT, whose engineered top-gate dielectric is tantalum pentoxide /silicon dioxide (Ta_2O_5/SiO_2), can obtain a high sensitivity as high as 649.04 mV/pH [13-16]. TiO_2 used as the top-gate dielectric also has been researched, which pronounced that TiO_2 thin film was able to increase the sensitivity amplification factor for the DG operation mode for its high dielectric constant of 80 [14]. In addition to TiO_2, the HfO_2 thin film has wide bandgap energy, large conduction band offset, and thermal stability [15]. C.-H. Lu et al. displayed high-sensitivity DG a-IGZO ISFETs with three HfO_2 TG gate dielectrics. Compared with the simple SG mode, the a-IGZO ISFET sensors in the DG mode show an extremely higher pH sensitivity of 1610mV/pH, which is far more than the Nernst limit [16]. Aside from those, several other high dielectric constant materials have been investigated. Particularly, the pH sensitivities of SiO_2, Silicon nitride (Si_3N_4), stannic oxide (SnO_2), HfO_2, zirconium dioxide (ZrO_2) and Ta_2O_5 were 356.1, 398.3, 426.0, 438.8, 470, and 478 mV/pH separately [17-20].

Summary： Recently, pH sensors with high sensitivity have attracted a great deal of attention due to their various merits. In this work, we have discussed the performance of pH sensors with different structures and materials based on the reported experimental data. Among the structures mentioned above, the triple-gate pH sensor shows an excellent sensitivity by utilizing the capacitive coupling with an extended gate detector. With regard to materials, the most popular active channel semiconductor is still the a-IGZO. In the dual-gated pH sensors or triple-gated pH sensors, in order to obtain a higher capacitive coupling ratio between the top-gate dielectric and bottom-gate dielectric, employing a high dielectric constant material Ta_2O_5 which is used frequently as the top gate dielectric is a good choice. In the ISFET pH sensors, CeO_2 thin films have several advantages as the gate dielectric. What's more, incorporate element into CeO_2 thin films can avoid the moisture absorption.

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