Transient Characteristics for Proton Gating in Laterally Coupled Indium-Zinc-Oxide Transistors (Postprint)

Authors: Liu, N, Zhu, LQ, Xiao, H, Wan, CJ, Liu, YH, Chao, JY

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Abstract

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Full Text

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Controlling and detecting the processing, transport, and delivery of chemical species is of great importance in sensors and biological systems. The transient characteristics of chemical species migration reflect fundamental properties of these processes. Here, we observed field-configurable proton effects in a laterally coupled transistor gated by phosphorosilicate glass (PSG). The bias applied to the lateral gate modulates the interplay between protons and electrons at the PSG/indium-zinc-oxide (IZO) channel interface. Due to the modulation of proton flux within the PSG films, the IZO channel current is correspondingly modified. The characteristic time for proton gating is estimated to be on the order of 20 ms. Such laterally coupled oxide-based transistors with proton gating are promising for low-cost portable biosensors and neuromorphic system applications.

Keywords: proton gating, laterally coupling, interfacial electric-double layer (EDL), transient characteristics

1. Introduction

In ion transistors, ions and charged molecules act as the charge carriers. Due to the fact that such carriers carry specific chemical information and functionality, they can selectively regulate various functions and processes. Thus, ion transistors can be useful for delivering, regulating, and establishing signaling patterns of ions and biomolecules in circuits. Furthermore, ions, molecules, or charged molecules can be selectively transferred using specifically modified nanopores/nanochannels in ion transistors. In these nanofluidic systems, ionic/electronic coupling occurs such that the gate can control and monitor the ionic currents. Ion migration can also be utilized to greatly modulate the carrier transport characteristics of condensed materials. In ionic liquid or ionic gel electrolyte-gated transistors, movements of anions/cations in the electrolyte toward the positively/negatively charged electrode form an electric-double-layer (EDL), composed of a compact Helmholtz layer and a diffuse layer. As a result of charge separation within a few angstroms, a high EDL capacitance above 1 F/cm^2 can be obtained. Therefore, field-induced electronic phase transitions and superconductivity are observed for condensed materials in transistor configurations. In these devices, the gate bias triggers ion migrations within the electrolyte, consequently inducing changes in carrier transport characteristics of condensed materials. Recently, wide band-gap oxide semiconductors have been reported to be gated by solid-state electrolytes, such as nanogranular SiO , nanogranular Al O , WOx, chitosan, etc. Though the proton conductivities for these electrolytes have been discussed, the transient characteristics for proton gating still need to be discussed in terms of characteristic time.

On the other hand, biological activities are always performed with ion signals rather than electrons. Therefore, it is highly desirable to translate ionic signals into easily detectable electronic signals. Thus, the realization of ionic/electronic coupled devices is meaningful for interfacing between electrical signals and biological response in biological systems. In this work, we observed field-configurable proton effects in a laterally coupled transistor configuration. Due to the modulation of proton flux within the nanogranular phosphorosilicate glass (PSG) films, the indium-zinc-oxide (IZO) channel current is correspondingly modified. The transient characteristics of the proton gating were discussed, resulting in the extraction of characteristic time for proton gating. The laterally coupled oxide-based transistors with proton gating are promising for low-cost portable biosensors and neuromorphic system applications.

2. Experimental Procedure

Laterally coupled IZO transistors were fabricated on glass substrates at room temperature, as schematically shown in [Figure 1: see original paper]. First, nanogranular phosphorosilicate glass (PSG) films were deposited on glass substrate and Si wafer at room temperature by plasma-enhanced chemical-vapor deposition (PECVD) using SiH (94% SiH + 6% PH) and O as reactive gases. For characterizing the proton conductivities, the PSG film was also deposited on ITO glass substrate. Scanning electron microscopy (SEM) characterizations indicate spongy microstructures for the as-deposited PSG films, as shown in the left panel of Figure 1. Next, patterned 150 nm thick IZO films were deposited on the PSG-coated glass substrate by radio-frequency (RF) magnetron sputtering with a nickel shadow mask. The sputtering was performed using an IZO ceramic target with an RF power of 100 W and a working pressure of 0.5 Pa. The size of the IZO patterns is 1000 × 150 m. The right panel of Figure 1 illustrates the top-view image of the IZO patterns by an optical microscope (Olympus DX51). A thin IZO layer will be self-assembled between two IZO patterns with a distance of 80 m, while no IZO layer will be self-assembled between two IZO patterns with a distance of 300 m, as schematically shown in Figure 1. When the isolated IZO pattern is deemed as the gate and the IZO patterns with self-assembled thin IZO layer are deemed as the source and drain electrodes, a laterally coupled IZO transistor is obtained. The thin IZO layer is deemed as the channel. The channel width (W) and length (L) were 1000 and 80 m, respectively. The isolated IZO patterns were also deposited on PSG-coated ITO conductive glass substrate to obtain an IZO/PSG/ITO capacitor.

Electrical characteristics were performed in a probe station in air ambient with a relative humidity of 60%. Samples were kept in a dark box to prevent interference from visible light. Proton conductivities and frequency-dependent capacitances of the PSG films were characterized by a Solartron 1260A impedance analyzer with an AC amplitude of 0.1 V and DC amplitude ranged between −0.5 and +1.0 V. Electrical performances of the laterally coupled IZO transistor were characterized by using Keithley.

3. Results and Discussion

Proton conductivities ( ) of the nanogranular PSG films were measured with an IZO/PSG/ITO capacitor, as shown in the inset of Figure 2a. A typical Cole-Cole plot is shown in Figure 2a with a DC bias of 0 V. The impedance spectroscopy data are collected as real (Re Z) and imaginary (Im Z) components of the complex impedance. The impedance real value (R) where the impedance imaginary value is zero is used to obtain the   value by using the following relation:

$$\delta = \frac{D}{A(R - R_0)}$$

where D, A, and R are the thickness of the PSG film, the electrode surface area, and the resistance of electrode, respectively. The thickness D is 1280 nm, A is 1.5 × 10^{3} cm^{2}, while R is 20 Ω. Thus,   is estimated to be 9.3 × 10   S/cm when DC bias is 0 V. The bias-dependent   values have also been studied, as shown in Figure 2b. It is observed that the   values increase slightly from 9.2 × 10   to 9.6 × 10   S/cm with the DC bias increasing from −0.5 to +1.0 V.

Frequency-dependent specific capacitances of the PSG films were measured with an in-plane sandwich structure, as shown in the inset of Figure 2c. Figure 2c illustrates a typical frequency-dependent specific capacitance measured with a

DC bias of 0.5 V. The frequency ranged from 1.0 Hz to 1.0 MHz. It is observed that the capacitance increases gradually with decreasing frequency. A maximum specific capacitance (C) of 2.0 F/cm^2 is obtained at 1 Hz, which is due to the formation of an EDL at the PSG/IZO interface. Under the external electric field, an oriented proton flux will be triggered within the PSG films by a sequence of proton hopping between hydroxyl groups and water molecules existing in nanogranular PSG films. Thus, protons accumulate at the PSG/IZO interface, resulting in a large EDL capacitance. The bias-dependent EDL capacitances have also been studied, as shown in Figure 2d. It is observed that the EDL capacitance is scattered between 1.8 and 2.1 F/cm^2 with the bias ranged between −0.5 and +1.0 V. Though proton fluxes are triggered by the external electric field, the EDL capacitance is not obviously affected by the bias applied on the PSG films.

Thus, in the configuration of lateral coupled transistors, the bias on the lateral gate would modulate the interplay between protons and electrons at the PSG/IZO channel interface. As a result, the carrier transport characteristics of the IZO channel will be modulated, as discussed below. The inset in Figure 2d shows the leakage current of PSG films measured on the in-plane sandwich structure. A low leakage current below 5 nA is obtained in the voltage range from −1.5 to +1.5 V, guaranteeing that the drain current (I_DS) will not be affected by the leakage current.

[Figure 3: see original paper] shows the typical output characteristics (I_DS vs V_DS) of the laterally coupled IZO transistor. V_DS is scanned from 0 to 1.5 V, while V_GS is increased from −0.6 to +1 V in steps of 0.2 V. The output characteristics of the IZO transistor in accumulation exhibit strong saturation and clear pinch-off. At low V_DS, I_DS increases linearly with V_DS, indicating that the device has good ohmic contact. Figure 3b shows the transfer characteristics (I_DS vs V_GS) of the device with a constant V_DS of 1.5 V. The sweep rate is 50 mV/s. A clear anticlockwise hysteresis window (ΔV_th) of 0.3 V is observed, which is likely due to the existence of mobile protons within the SiO proton conductor. The current on/off ratio is estimated to be 5 × 10 . The results indicate that the gate bias triggers proton flux within the PSG layers, obviously modulating the IZO channel conductivities. A low subthreshold swing of 110 mV/decade is observed on the laterally coupled IZO transistor. A threshold voltage (V_th) of −0.4 V is estimated from the I_DS^(1/2) vs V_GS plot, indicating that the device operates in depletion mode. Field-effect mobility (_FE) in the saturation region can be obtained by the following equation:

$$\mu_{FE} = \frac{2L}{WC_0}\left(\frac{\partial\sqrt{I_{DS}}}{\partial V_{GS}}\right)^2$$

where W is the channel width of 1000 m and L is the channel length of 80 m, whereas C is the EDL capacitance of 2.0 F/cm^2.

The V_GS-dependent _FE is shown in Figure 3c. A maximum mobility of 24 cm^2 V ^{1} s ^{1} is obtained. To investigate the interface state density (N_it) at the PSG/IZO channel interface, the transfer curve of the lateral coupled TFT was measured in the linear region with a fixed V_DS of 0.2 V, as shown in Figure 3d. The subthreshold swing (S) is estimated to be 89 mV/decade. The interface state density (N_it) can be estimated by the following equation:

$$N_{it} = \left(\frac{S \cdot \log(e)}{kT/q} - 1 \right) \frac{C_0}{q}$$

where k is Boltzmann’ s constant, T is the absolute temperature (300 K), and q is the electron charge. N_it is estimated to be 6.1 × 10^{12} cm ^{2}, which is comparable to that for high-k oxide/semiconductor interfaces. The results discussed above indicate that proton gating modulates the channel conductance significantly.

Transient characteristics can reflect the dynamic properties of proton gating. Moreover, the characteristic time as an important parameter for proton gating is also meaningful for interfacing between electrical signals and biological response in biological systems. Here, transient channel currents induced by gate pulses through proton gating are recorded. Figure 4a [Figure 4: see original paper] shows the transient I_DS of the laterally coupled IZO transistor measured with a pulsed V_GS (1 V, 10 ms) under a constant V_DS of 0.2 V. At the end of the gate pulse, a peak I_DS of 21.6 A is obtained. When the gate pulse is finished, I_DS gradually decays back to the resting current of 15 A in 500 ms. The lateral migration of protons within PSG films plays an important role in the transient I_DS. When a positive pulse voltage is applied on the lateral gate, protons migrate and accumulate at the PSG/IZO channel interface, thus a high I_DS is obtained, as shown in the top-left panel of Figure 4a. When the pulse voltage is finished, protons gradually drift back to their equilibrium positions. Correspondingly, I_DS decreases gradually back to the resting current. It was reported that hydrogen diffusion in a-Si, defect diffusion, oxygen vacancy diffusion, and ion hopping transport in complex condensed-matter systems agreed well with a stretched exponential model. Similarly, the movements of protons within nanogranular PSG films are also responsible for the decay process of I_DS. Thus, I_DS decay curves can be fitted by a stretched exponential function (SEF) shown below:

$$I_{DS}(t) = I_\infty + (I_0 - I_\infty) \exp \left[- \left(\frac{t - t_0}{\tau} \right)^\beta \right]$$

where   is the retention time, t is the time when the pulse voltage is finished, I is the triggered current at the end of the pulse voltage, I_∞ is the resting current, and   is the stretch index ranging between 0 and 1. A good fitting curve for the I_DS decay triggered by the gate pulse is obtained, as shown in Figure 4a (red line). The retention time ( ) is estimated to be 17.8 ms.

When several positive gate pulses (1 V, 10 ms) are applied to the laterally coupled IZO transistor, repeated transient I_DS are observed, as shown in Figure 4b. Each transient I_DS is also fitted with the equation above. The obtained   values are shown in Figure 4c with high stability.

The transient I_DS induced by negative gate pulses through proton gating are also recorded. When several negative gate pulses (−1 V, 10 ms) are applied to the laterally coupled IZO transistor, repeated transient I_DS are also observed. The transient I_DS are fitted with the equation as well. Stable retention times ( ) of 22 ms are obtained, as illustrated in Figure 5a [Figure 5: see original paper]. The retention time is larger for negative gate pulses than for positive gate pulses. These slight differences can be explained as follows. The positive gate pulse results in accumulation of protons at the IZO channel/PSG interface. When the gate pulse ends, protons diffuse back; the positive drain bias promotes such diffusion. Thus, the retention time is smaller. Conversely, the negative gate pulse results in accumulation of protons at the gate/PSG interface. When the gate pulse ends, the positive drain bias restrains the back diffusion of protons. Thus, the retention time is larger.

The transient I_DS are also triggered by positive pulses (1 V, 10 ms) and negative pulses (−1 V, 10 ms) in a sequential mode. The retention times for each pulse are also obtained, as shown in Figure 5b. The retention time is always 18 ms for positive gate pulses and always 21 ms for negative gate pulses. Thus, the positive pulses and negative pulses do not affect the transient characteristics of each other. Moreover, it can also be concluded that the intrinsic properties for proton gating are not significantly affected by gate pulses in the range between −1 and +1 V. The stable retention time also suggests that the proton gating is repeatable without electrochemical reaction at the interface. It is interesting to note here that the characteristic time for neural activity is tens of milliseconds or hundreds of milliseconds; thus, the proton-gated laterally coupled IZO transistors are also interesting for neuromorphic system applications.

4. Conclusion

In summary, laterally coupled IZO transistors gated by nanogranular PSG films were fabricated on glass substrates at room temperature. Due to the modulation of the lateral proton flux within the PSG films, the IZO channel current is correspondingly modified. Good transistor performances are obtained, including a low operation voltage of 1.5 V, a low subthreshold swing of 110 mV/decade, a high current on/off ratio of 5 × 10 , and a high _FE of 24 cm^2 V^1 s^1. With the transient channel current triggered by gate pulses, the characteristic time for proton gating is estimated to be 20 ms. Due to the similar characteristic time scale between the proton gating in the laterally coupled oxide transistors and neural activities in biological systems, the proton-gated devices proposed here are interesting for portable biosensors and neuromorphic system applications.

AUTHOR INFORMATION

Corresponding Author *L. Q. Zhu. E-mail: lqzhu@nimte.ac.cn.

Notes The authors declare no competing financial interest.

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