Roll-to-Roll processable OTFT-based Amplifier and Application for pH sensing

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***Abstract*—The prospect of roll-to-roll (R2R) processable Organic Thin Film Transistors (OTFTs) and circuits has attracted attention due to their mechanical flexibility and low cost of manufacture. This work will present a flexible electronics application for pH sensing with flexible and wearable signal processing circuits. A transimpedance amplifier was designed and fabricated on a polyethylene naphthalate (PEN) substrate prototype sheet that consists of 54 transistors. Different types and current ratios of current mirrors were initially created and then a suitable simple 1:3 current mirror (200nA) was selected to present the best performance of the proposed OTFT based transimpedance amplifier (TIA). Finally, this transimpedance amplifier was connected to a customized needle-based pH sensor that was induced as microfluidic collector for potential disease diagnosis and healthcare monitoring.**

Keywords—OTFT, R2R, pH, Flexible electronics, Transimpedance amplifier

I. INTRODUCTION

Over the past decade, a variety of flexible organic sensors have been created with different architectures to detect characteristic signals from the human body, such as temperature and arterial pulses. Organic Thin Film Transistor (OTFT)-based sensors had been shown capable to detect pH, ionic concentration, and protein from body fluids with different selective materials on semiconductor or an extended gate [1-5].

For the development of wearable devices, sensors and signal conditioning circuits are two important aspects in practical manufacture and application. For most OTFT sensors, signals directly generated by a sensing reaction, such as ion-induced charge redistribution and electrochemical reaction, are relatively low, i.e. in the range of 1μA or lower, and the signal cannot be output unless firstly amplified. Experimentally, the signal can be extracted with a bulk device that is not flexible and wearable. But for flexible application, it is necessary to create suitable signal-processing circuits on a flexible substrate to transduce or amplify the signal generated on wearable sensor to an externally readable level [6].

Roll-to-Roll processing is very important for the low-cost manufacture of OTFTs with high throughput [7]. Our approach uses deposition methods that are high rate and room temperature, without annealing or solvent evaporation steps to facilitate R2R processing. In previous research, a flash evaporation technique was used to deposit polymer thin film on fast moving plastic webs, enabling deposition of polymeric dielectric materials for OTFT at high speed. Recently, small molecule semiconductors have got attention for OTFTs, for example dinaphtho[2,3-b:2',3'-f] thieno[3,2-b]thiophene (DNTT) exhibits higher stability than pentacene and P3HT. In addition, extra oxygen-doping enables relatively high mobility [8-11].

Although a single transistor can be used as an amplifier, the actual amplifying effect is mainly determined by materials properties of the transistor, such as semiconductor mobility, doping concentration and dielectric relative permittivity. A single OTFT can amplify an applied voltage signal at the gate to generate a current signal between source and drain (voltage control current source-VCCS). To achieve a broader amplification range and signal converting method, an integrated circuit can be constructed with several sub-circuits and pseudo resistors which were created from transistors.

A current mirror (CM) is a basic subcircuit of integrated circuits design that replicates single or multiple output currents for supplying other subcircuits with a single fixed current source at the initial stage (current range can be fixed with a resistor or pseudo resistor). It is often used as a bias unit in a circuit to provide a known current to an analogue circuit [12]. A transimpedance amplifier is often defined as a current-to-voltage (I – V) converter. In recent research, differential or transimpedance amplifiers have been fabricated with solution-based methods on flexible substrates, with working voltage in the range from -5V to 5V, with voltage gain in the range from 1 to 50 [13-15].

This research work applies OTFTs manufactured with an all-vacuum evaporation process. OTFT-based current source and TIA were made by evaporating copper interconnects, and then circuits were validated with a Keithley 2400 source meter and oscilloscope to ensure the linearity of input current and output voltage of the TIA. Finally, a pH sensing unit which included a customized needle-based pH sensor (voltammetric sensor) in conjunction with a commercialized buffer amplifier and a resistor (1 kΩ) was connected to the flexible circuit to extract a signal of pH variance. A needle type microdialysis system has used to perform body-worn sweat lactate and glucose measurements during exercise [16]. This research work uses similar approach with pH sweat sensor[17] and builds a sensing system to extract a signal from buffer solutions with different pH value that is used to mimic human sweats.

II. Experiments

A. Fabrication of OTFTs

The 100 mm x 100 mm PEN substrates for OTFTs were pre-cleaned with isopropyl alcohol (IPA) and deionized water. The metallization of gate electrodes was achieved by thermally evaporating Al to a thickness of 50 nm through a shadow mask. Afterwards, the metallized substrate was fixed on the drum of a R2R vacuum web-coater and rotated at a linear speed of $25\ m/min$ to deposit a thin layer of Tripropylene Glycol Diacrylate (TPGDA) dielectric polymer ($650\ nm$) by flash evaporation. TPGDA monomer was initially vaporized in a hot tank (270 ^{o}C), and then deposited via a heated nozzle to condense as a liquid onto the cooler substrate. The deposited monomer was cured under Ar plasma that initiated the cross-linking reaction of the monomer. Afterward, a thin layer of polystyrene was spin-coated from toluene solution onto the surface of the TPGDA to deactivate the polarity of surface, on which, a thin layer of organic semiconductor – dinaphtho [2,3-b:2',3'-f] thieno[3,2-b]thiophene (DNTT) –was evaporated at 0.01 nm/s. Finally, a layer of gold was evaporated through a brass shadow mask onto the DNTT surface as contact electrodes. After fabrication, all OTFTs are stored in a dark environment at approximately 20 ^{o}C and 5% – 10% humidity for 96 hours for stabilization.

B. Circuit layout design

The initial circuit included three parts consisting of nine transistors with different widths. For the first part, a simple current mirror, a common gate is used to connect transistors in parallel and minimize the space of the layout. Similar geometry of layout was applied to multiple widths to facilitate tuning width/length for circuit design. The second part is an input transistor with higher width/length ratio (*W/L*), therefore, multiple transistors with wider channel are placed in parallel, so that the actual width can be tuned by combining connections as well. Transistors in the third part were used as a pseudo resistor to regulate the overall impedance of the circuit. The layout scheme is shown in Figure 1. We manufactured 54 transistors in one batch, and transistors with similar properties were selected to fabricate the functional circuit. The *W/L* ratio of transistors in the top region and bottom region are $2.5mm/0.1mm$ and that of transistors in middle region is $25mm/0.1mm$. On the top left or top right region, any two or more transistors with similar properties were used to fabricate a current mirror with different current transfer ratios. In the middle region, the 18 transistors with longer channel width are available for the input transistor cluster, and in each of the bottom left and bottom right regions, there are 9 transistors available for fabrication of pseudo resistors to tune the overall impedance of the entire circuit. The common gate design was used to reduce the extra gate-to-gate interconnections required when fabricating current mirrors and the impedance-tuner.

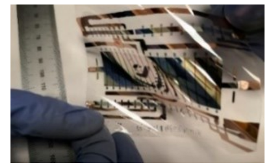

Fig. 1. All-evaporation based circuit on flexible substrate (54 Transistors in one batch and only 21 transistors are used to build signal conditioning circuit)

C. Fabrication and Characterisation of Current Mirror and Amplifier

Initially, a series of single gate transistors were used to fabricate current mirrors (Figure 2) to test their performance. Because the current mirror is to supply current to a differential pair, selecting an appropriate number of transistors on the reference side and mirror side, i.e. tuning channel W/L ratio on each side is helpful for designing the transimpedance converter. Transistors in the current mirror (T1, T2 and T3 in Figure 3) work in the saturation region which follow (2) and transistors of differential pair (T4 and T5 in Figure 3) work in the linear region which follows (1),

$$I_{ds,lin} = \frac{W}{L}\mu C_i\left(V_g - V_T\right)V_{ds} \quad (1)$$

$$I_{ds,sat} = \frac{W}{2L}\mu C_i\left(V_g - V_T\ \right)^2 \quad (2)$$

where $I_{ds,lin}$ and $I_{ds,sat}$ are drain-source current in the linear region and saturation region respectively, W and L are width and length of channel, μ is mobility, C_i is the capacitance per unit area, V_T is threshold voltage, V_g and V_{ds} are gate voltage and drain-source voltage respectively. Transistors are connected by an evaporated copper trace through a shadow mask.

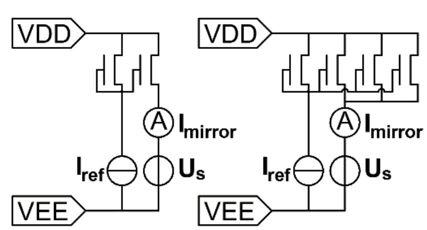

Fig. 2. Current Mirror fabricated by transistors with same width/length ratio (Left: 1:1 CM and Right: 1:3 CM)

Current mirrors were tested with fixed *VDD* (10 V) and *VEE* (0 V). A voltage source (U_s) was applied to sweep a voltage from 10 V to 0 V in order to vary (I_s), from 0.1 µA to 1 µA. The measured current was plotted against input voltage. After that, the current transfer ratio (I_o/I_r) is calculated under different U_s and plotted against the reference current I_r.

The current transfer ratio is an important parameter to evaluate performance of a current mirror, shown in (3).

$$M = \frac{I_{output}}{I_{reference}} = \frac{(W/L)_{mirror}}{(W/L)_{reference}} \quad (3)$$

Next, a batch of common gate transistors with a pre-designed pattern is created to make a transimpedance amplifier. Interconnects are created in this case by thermally evaporating copper through a shadow mask and conductive silver paste was applied on the surface of the electrode to make a convenient and robust connection with external wires. The *W/L* ratios of the amplifier are shown in Table I.

Table 1. W/L Ratio of each transistor

Transistor	W/L Ratio
T1	2500 µm/100 µm
T2	7500 µm/100 µm
T3	7500 µm/100 µm
T4	25000 µm/100 µm
T5	25000 µm/100 µm
T6	7500 µm/100 µm
T7	7500 µm/100 µm
T8	12500 µm/100 µm
T9	2500 µm/100 µm

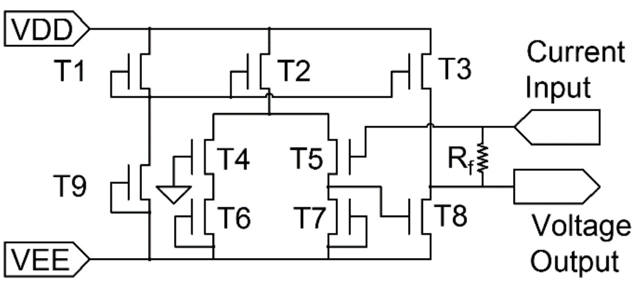

Fig. 3. Circuit of transimpedance amplifier

D. System Integration of pH Sensing

A needle electrode is functionalized with hydrogen ion selective materials as the method described in [17]. The hydrogen ion selective membrane enables a potential difference between working electrode and reference electrode when the needle sensor was immersed into solution. In order to avoid an impedance match problem and to convert the potential that generated from the sensor to a current source for testing the transimpedance amplifier, a voltage buffer (LMP7721, Texas Instruments) was needed. A 1 kΩ resistor was connected in series between the output of the buffer amplifier and one of the inputs of the flexible TIA. Finally, the output voltage - V_{out} of the transimpedance amplifier was measured with an oscilloscope (Figure 4).

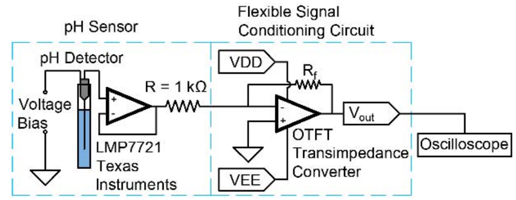

Fig. 4. Schematic of pH sensing system

III. Result and Discussion

A. Characterisation of Current Mirror and Transimpedance Amplifier

M is determined by the ratio of the collective W/L of transistors at the reference side and transistors at the mirror side. Because all single gate transistors are designed to have the same geometry, *M* should be determined by the actual channel width on each side, i.e. the number of transistor on each side. A series of current mirrors with different number ratio (1:1 and 1:3) were made and *M* calculated. Figure 5 shows *M* for a 1:1 and 1:3 current mirror. It's noticeable that measured *M* of current mirror does not match the actual number ratio; this might be induced by the geometrical mismatch caused by manufacturing, non-uniform deposition of organic semiconductor and different contact resistance of transistors. In order to obtain a positive voltage readout for future low power wearable system at the output stage of the amplifier, simple current mirrors of 1:3:3 (channel width ratio of T1:T2:T3) were selected in the design of this transimpedance amplifier.

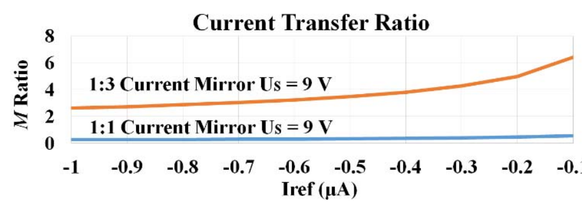

Fig. 5. DC current transfer ratio for 1:1 and 1:3 current mirror

The transimpedance amplifier was used to convert a small current signal to a voltage signal, by connecting a resistor between the input end and output end of the amplifier. In this work, a resistor (R_f) of 820 kΩ was used as gain setting, shown in Figure 3. The input current ranged from 0 to 3 µA in 0.1 µA steps, and the output voltage measured by oscilloscope, is shown to vary linearly with input current with a small voltage offset, shown in Figure 6. After regression analysis on this line, the experimental gain was calculated (1.1 MΩ). The difference between theoretical value and practical value might be induced by the impedance mismatch of transistors.

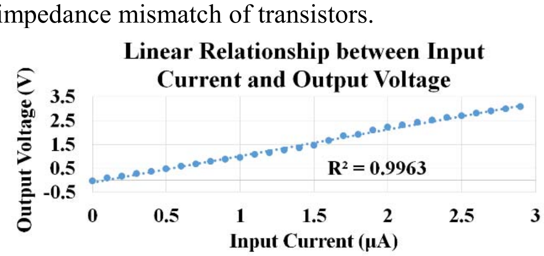

Fig. 6. Output voltage from the trans-impedance amplifier showing linear relationship between input current and output voltage (R_f= 820 kΩ)

B. *pH sensing and signal conditioning*

After obtaining the linear relationship between input current and output voltage on the flexible transimpedance amplifier, a pH sensing test was set up. Shown in figure 4, the pH sensor consist of a pH detector and a buffer amplifier. A bias voltage of 0.87 V at the reference electrode was applied during the test. The potential changes from pH sensor were firstly processed by a voltage buffer and a resistor of 1 kΩ. The current changes were converted to voltages through the TIA, and Figure 7(a) displays the output voltage of the TIA for measuring different pH (4, 6, 7 and 9) levels. There is a noticeable step-wise increase in output voltage with stepwise increases in pH value. Figure 7(b) shows the relationship between output voltage and pH.

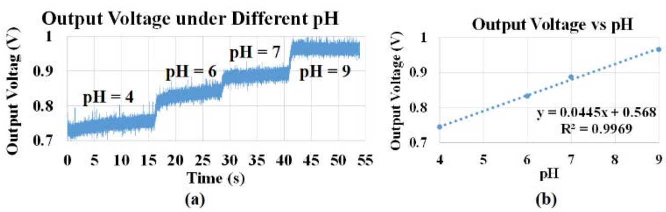

Fig. 7. (a) Real time measurement of pH (b) A linear relationship between pH and Output Voltage

The results show that the OTFT-based TIA manufactured by roll-to-roll process is capable to distinguish different levels of pH in buffer solution. In this research work we aimed to implement a system (sensor and signal conditioning integrated circuit) that can sense human sweat from pH levels from 4 to 7.

IV. Conclusion

In this work, flexible OTFT sheets were manufactured using the Oxford webcoater, which is capable of Roll-to-Roll processing. Based on this technique, a series of current mirrors and a transimpedance amplifier were all manufactured using high-rate deposition techniques on a flexible substrate, although the sample tested here is 10 x 10 cm^2 in order to provide a flexible platform for device testing, an integrated design based on this successful circuit has now been designed with a reduced size (4 x 4 cm^2), a thin layer of PDMS will be applied as protective film as well as a biocompatible encapsulation for a body-worn device. Future work will focus on designing an OTFT-based buffer amplifier to replace the commercial buffer amplifier, depositing the pH sensor directly onto the flexible electrode manufactured by thermal evaporation via Roll-to-Roll process, testing the mechanical flexibility of working circuits and testing on healthy subjects.

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