Organic Electronics 56 (2018) 170–177

Contents lists available at ScienceDirect

Organic Electronics

journal homepage: www.elsevier.com/locate/orgel

Multifunctional sensor based on organic field-effect transistor and ferroelectric poly(vinylidene fluoride trifluoroethylene)

Stuart Hannah[a], Alan Davidson[a], Ivan Glesk[a], Deepak Uttamchandani[a], Ravinder Dahiya[b], Helena Gleskova[a,*]

[a] Department of Electronic and Electrical Engineering, University of Strathclyde, Glasgow, G1 1XW, United Kingdom
[b] School of Engineering, University of Glasgow, James Watt South Building, Glasgow G12 8QQ, United Kingdom

ARTICLE INFO

Keywords:
Sensor
Capacitance
P(VDF-TrFE)
Organic transistor
Force
Temperature

ABSTRACT

A multifunctional sensor that responds to all – static/quasi-static or dynamic temperature or force – is reported. The sensor is based on a ferroelectric poly(vinylidene fluoride trifluoroethylene) (P(VDF-TrFE)) capacitor connected to the gate of organic field-effect transistor (OFET). Both, the P(VDF-TrFE) capacitance and the output voltage of the P(VDF-TrFE)/OFET sensor exhibit a logarithmic response to static compressive force, leading to higher sensitivity for small forces. In addition, both the P(VDF-TrFE) capacitance and the output voltage of the P(VDF-TrFE)/OFET sensor exhibit a linear dependence on the static/constant temperature. Response to static force or temperature is observed irrespective of whether P(VDF-TrFE) is in ferroelectric or paraelectric states, confirming that piezo/pyroelectricity is not essential when monitoring static events. The piezo/pyroelectricity become activated during dynamic events (dynamic force or temperature) when the ferroelectric P(VDF-TrFE)/OFET sensor is used. The obtained results indicate different sensing mechanisms for static and dynamic stimuli. Consequently, by choosing P(VDF-TrFE) layers in ferroelectric or paraelectric states a route for differentiating between the static and dynamic stimuli may exist.

1. Introduction

The field of tactile sensing has been growing rapidly in recent years and applications span from touch screen interfaces to electronic skin (e-skin) for humanoid robots and electronic textiles (e-textiles). An e-skin is expected to replicate the complex sensory response of real skin, e.g. pressure and temperature transduction, and also possess the ability to stretch and conform to non-planar surfaces [1]. Consequently the demand for multimodal sensing (sensing of various stimuli) and surface mapping (number of sensors per unit area) favors sensor arrays, i.e. arrays of pixels that contain sensing devices for targeted external stimuli. On the other hand, discrete unobtrusive sensors placed in garments at specific body locations are suitable for electronic textile applications. Examples include physiological sensors (sensing body's physiological state), bio-kinetic sensors (tracking individual's movement), and ambient sensors (impact of the environment on the individual).

Thin-film sensors for pressure/force/strain or temperature have been the focus of many recent research activities aimed at e-skin or e-textiles. For pressure/force/strain detection they exploit the piezoresistive effect [2–4], the piezoelectric effect [5–8] or capacitive changes [9,10], whereas for temperature sensing capacitive [11] and pyroelectric-based sensors [12] can be found.

To harvest the response of certain materials to pressure/force/strain or temperature stimuli, the sensors often contain field-effect transistors (FETs) that convert the response generated by the material to an amplified voltage signal, suitable for subsequent interfacing with readout electronics [4,7,8,13–19]. Here, ferroelectric polymers are very attractive because they exhibit piezo- and pyro-electric properties, while being inherently compatible with plastic substrates. Some examples of previous work include a polyvinylidene difluoride (PVDF) capacitor combined with an organic charge-modulated FET for the detection of dynamic force [13] or changing temperature [14], a poly(vinylidene fluoride trifluoroethylene) (P(VDF-TrFE)) capacitor combined with an organic electrochemical transistor for contactless touch detection [15], or P(VDF-TrFE) layer integrated with a CMOS FET to form a piezoelectric oxide semiconductor field-effect transistor (POSFET) for touch sensing [17]. In these devices, the piezoelectric or pyroelectric properties of the ferroelectric polymers have been exploited, allowing sensing of the dynamic/changing force or temperature. A human skin, however, is able to detect static/constant as well as dynamic/changing stimuli, and distinguish between them. Temperature measurements

* Corresponding author. Department of Electronic and Electrical Engineering, University of Strathclyde, Royal College, 204 George Street, Glasgow, G1 1XW, United Kingdom.
E-mail address: helena.gleskova@strath.ac.uk (H. Gleskova).

https://doi.org/10.1016/j.orgel.2018.01.041
Received 23 May 2017; Received in revised form 29 December 2017; Accepted 30 January 2018
Available online 11 February 2018
1566-1199/

may also involve detection of static/constant stimuli since temperature changes may be occurring on long time scales. Therefore, it would be desirable if the sensors utilizing P(VDF-TrFE) would provide the measurement of both the static and dynamic stimuli.

Previously, we have reported that the capacitance of a ferroelectric P(VDF-TrFE) layer changes with force or temperature [20]. In addition, the ferroelectric co-polymer exhibits piezo- and pyroelectric properties. In this paper we present a sensor based on a P(VDF-TrFE) capacitor and a low-voltage organic field-effect transistor that responds to static/quasi-static force or temperature. Having the ability to also respond to dynamic force or temperature, such sensor offers the desired multi-functionality. The use of one single material, such as P(VDF-TrFE), also lends a major advantage during the fabrication.

2. Experimental procedures

Parallel-plate capacitors containing a 2.5-μm-thick P(VDF-TrFE) (Piezotech) layer sandwiched between two metal layers (Al and Au) were fabricated using the procedure described previously [21]. The crystalline structure, and consequently the piezoelectric and pyroelectric properties of the polymer film, depend on the molecular proportion x ($0.6 < x < 0.85$) of vinyledene fluoride in P(VDF$_x$-TrFE$_{1-x}$). Among various compositions of P(VDF-TrFE), the one with 65/35 wt ratio exhibits good ferroelectric response [22]. For this reason, P(VDF-TrFE) with 65/35 wt ratio was used in this work. A 10% P(VDF-TrFE) solution was spin coated at 3000 rpm for 30 s. Afterwards the polymer films were annealed to enhance the crystallization, to evaporate any left-over solvent and to remove any local stress generated during deposition. The films were annealed close to the melting point of P(VDF-TrFE), i.e. the temperature was ramped to 155 °C for 1 h, kept at 155 °C for 10 min, and then gradually lowered to room temperature in 4 h to minimise the stress. After the annealing, the polymer films were treated with hexamethyldisilazane (HMDS) followed by vacuum deposition of a 200-nm-thick top metal layer composed of thin Cr and thick Au. After fabrication, the capacitor was subjected to DC poling to induce the ferroelectric phase and subsequently mounted onto a glass slide for compatibility with the force testing equipment. Poling of P(VDF-TrFE) was achieved by applying a high electric field (100 V/μm) across its thickness in multiple steps as explained in Ref. [21]. The relative permittivity of such P(VDF-TrFE) was about 10.

Bottom-gate, top-contact p-channel OFETs based on dinaphtho[2,3-b:2′,3′-*f*]thieno[3,2-*b*]thiophene (DNTT) (Sigma Aldrich) were fabricated according to the procedure described in Ref. [23]. The bi-layer gate dielectric consists of aluminum oxide (AlO_x) prepared by UV/ozone oxidation and an alkyl phosphonic acid (C_nPA) (Strem Chemicals) monolayer prepared in vacuum. Transistors with $C_{14}PA$, $C_{16}PA$, and $C_{18}PA$ have been used, thereby allowing us to test the sensor functionality for a range of field-effect mobilities and threshold voltages [23]. The mobility of the transistors used in this study varied between 0.02 and 0.56 cm^2/Vs and the threshold voltage between −0.19 and −0.68 V. Fig. 1(a) shows the Al/AlO_x/C_nPA/DNTT/Au transistor structure. The transistors had nominal channel length of 30 μm and a channel width of 1 mm.

To finalize the sensor, P(VDF-TrFE) capacitor was connected to OFET gate electrode by gold wire. Unless otherwise noted, the Al electrode of P(VDF-TrFE) was connected to the gate. Fig. 1(b) shows the cross-section of the P(VDF-TrFE) capacitor with an area of 5 by 5 mm^2 and Fig. 1(c) presents the circuit diagram of the P(VDF-TrFE)/OFET sensor. To take advantage of the low-voltage transistor, the voltages were kept as low as possible. The drain (V_D) was always biased at −2 V. V_{IN} was chosen just high enough to allow the transistor to operate in saturation. The output voltage V_{OUT} was measured across the 1 MΩ resistor. In some instances, the voltage on the gate of the transistor V_G was also measured. The sensor measurements were performed in ambient atmosphere and therefore, the OFET transfer characteristics were also recorded under ambient light.

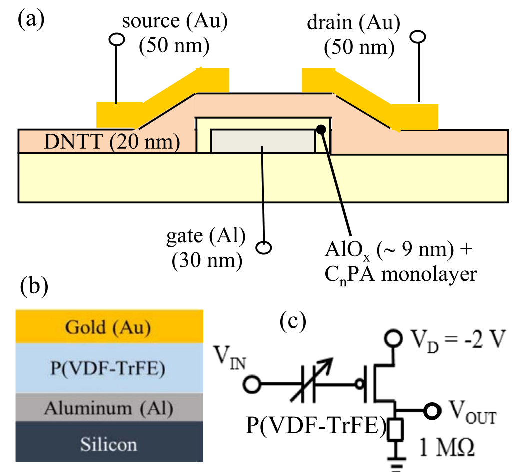

Fig. 1. Cross-sectional view of OFET (a) and P(VDF-TrFE) capacitor (b); schematic of the P(VDF-TrFE)/OFET sensor (c).

Static compressive force of up to ~9 N was applied in the normal direction to the P(VDF-TrFE) capacitor. Force was applied using a linear stage controlled by a stepper motor (Zaber Technologies) fitted with an in-house designed Teflon probe capable of compressing 40% of the capacitor's active area. Force was recorded with a load cell (Omega) and extracted after it had stabilized. All experiments were performed at room temperature. The minimum and maximum applied force was ~80 mN and ~9 N, respectively. Capacitance of a stand-alone P(VDF-TrFE) capacitor and V_{OUT} of a P(VDF-TrFE)/OFET sensor were measured as functions of the applied force. The output voltage V_{OUT} was referenced to the value of $V_{OUT} = V_{REF}$ with no force applied. $|\Delta V_{OUT}| = |V_{OUT} - V_{REF}|$ was calculated and plotted as a function of force. Measurements were also performed on a P(VDF-TrFE)/OFET sensor where the P(VDF-TrFE) capacitor was depoled by heating at 125 °C for 30 min (T_{Curie} ~ 110 °C) to induce a paraelectric state. The disappearance of the ferroelectric phase, i.e. piezoelectric and pyroelectric properties, was verified with a charge amplifier.

Similarly to the static force experiments, the stand-alone P(VDF-TrFE) capacitor and P(VDF-TrFE)/OFET sensor were studied as a function of P(VDF-TrFE) temperature. The temperature was applied to P(VDF-TrFE) capacitor via a Peltier element that was connected to a custom designed PID controller and LabView program. Temperatures in the range from ~20 to ~50 °C were used. No force was applied to P(VDF-TrFE) during the temperature experiments.

The electrical measurements were performed with Agilent B1500A semiconductor parameter analyzer equipped with a capacitance module. Capacitance was measured as a function of frequency and extracted at 2 kHz.

3. Results

3.1. Response of P(VDF-TrFE)/OFET sensor to static force

Fig. 2(a) shows the change in P(VDF-TrFE) capacitance when compressive static force is applied in the normal direction to a poled P(VDF-TrFE) capacitor. The capacitance is 858.6 pF when no force is applied. It rises with the increasing applied force and reaches a value of 886.9 pF at 8.85 N. The capacitance increases logarithmically, resulting in a larger change for low forces. If one defines the sensitivity as a change in capacitance per unit force, a sensitivity of ~7.5 pF/N is observed in the range from 0 to ~0.5 N. Fig. 2(b) shows the response of P(VDF-TrFE)/OFET sensor when a static force of 0.74 N lasting ~ 7 s is repeatedly applied to P(VDF-TrFE). Both the transistor drain current I_D and V_{OUT} are shown. Application of 0.74 N leads to a change in V_{OUT} of