Title	低電圧デュアルゲート有機トランジスタを用いた圧力検出素子に関する研究
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論文の内容の要旨

Part 1: Research Content

Background

An unprecedented drive to create a smart society based on the connectivity of electronic sensors to improve safety and quality of life has led to research into low-cost, flexible, and printable transistors. Billions of sensors are required to achieve this goal. Therefore, organic materials are good candidates to meet this demand of transistor switches for sensing applications because they require solution-processing methods to fabricate these devices known today as organic field-effect transistors (OFETs) at temperatures less or equal to 100°C on a wide range of substrates. Some of the advantages of using OFETs as readout elements for pressing sensing applications include monitoring of environmental pressure, electronic skin, human-machine interaction, monitoring of the footprints of the elderly in care homes [1-4]. Prototypes of organic pressure sensors reported operate in high voltages usually above 10 V; therefore, a need to develop organic pressure sensors that can be powered by portable battery cells is necessary. The operation voltage of the sensor is strongly dependent on the drive voltage of the OFET. Recently, our group demonstrated a pressure-sensing device using a piezoelectric copolymer film of Polyvinylidene Fluoride-Trifluoroethylene (P(VDF-TrFE)) as the sensing layer and a low-voltage OFET as the readout element [5]. The device configuration is such that pressure exerted on the sensing layer is assumed to induce the generation of charges from the piezoelectric layer, which transduces as threshold voltage and drain current changes observed in the electrical output of the OFET. However, the reason for the change in threshold voltage and drain current when pressure is applied to the sensing layer has not been quantitatively proved yet.

Objective

The objective of this research is to clarify the operation mechanism of the device (dual-gate organic pressure sensor) by quantitative analysis to determine the piezoelectric constant, d_{33} of the sensing layer was carried out. First, by reproducing the dual-gate organic pressure sensor device already reported [5]. In addition, proposing a sensing mechanism to describe the operation of the device. Second, by quantifying charges per unit area generated by the sensing layer using a conventional dual-gate OFET with CYTOP as the top-gate dielectric layer. Third, by estimating d_{33} from results of the dual-gate OFET and dual-gate organic pressure sensor devices, then comparing the result with that obtained by direct measurement using a quasistatic (belincourt) method.

Summary of results

Using dielectrics such as poly (vinyl cinnamate) (PVCN) and an environmentally friendly and degradable biopolyamide, low voltage OFET devices were fabricated and characterized in **chapter 2**. Photochemical crosslinking process was carried out to make PVCN insulator chemically resistant to chlorobenzene solvent while the biopolyamide material was characterized to confirm if it was chemically resistant to chlorobenzene**.** Using these insulators, the OFETs operated in a low voltage range, 1 V to -5 V, because of the reduced interface traps the dielectrics formed with the TIPS-Pentacene/polystyrene semiconducting blend. The few trap states allowed for charge carriers to quickly fill up the sub-gap density of states thus leading to a small subthreshold slope of values 108 mV/decade for the biopolyamide based OFET and 110 mV/decade for the PVCN based OFET. Figure 2 (a) and (b) shows the transfer characteristics of the PVCN and biopolyamide based OFET. The mobility of the OFETs was 0.27 cm^2/V.s and 0.18 cm^2/V.s for the biopolyamide and PVCN respectively. The results obtained using the biopolyamide increases the prospects of eco-friendly organic transistors available for use in the nearest future.

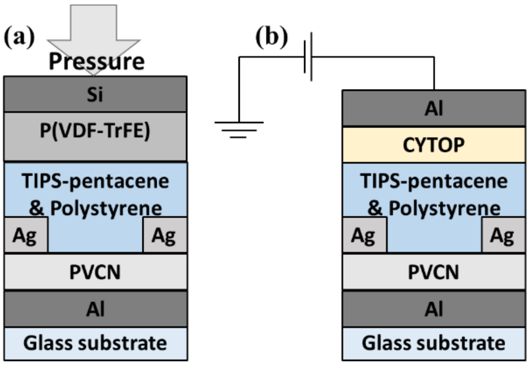

Fig. 1. Device structure of (a) low-voltage dual-gate organic pressure sensor (b) dual-gate OFET

In chapter 3, a dual-gate organic pressure sensor device was successfully fabricated using a polarized P(VDF-TrFE) layer. When the sensing layer (see Fig. 3(a)) is placed on the semiconducting layer of the low-voltage OFET, an initial shift in transfer characteristics is observed, thus confirming the polarization of the P(VDF-TrFE) layer. When pressure is applied on the piezoelectric layer, a corresponding shift in transfer characteristics to the left (see Fig. 3 (b)). Figure 3c shows the linear relation between pressures applied to the threshold voltage shift. The shift in transfer characteristics is similar to that of the conventional dual-gate OFET, just that in this case the sensing voltage generated by the deformed piezoelectric layer, induced the threshold voltage (V_{th}) shift observed. Therefore, **in chapter 4**, a low-voltage dual-gate OFET using CYTOP as top-gate dielectric was successfully fabricated. The top-gate voltage controlled was used to control V_{th} of the bottom gate potential [6].

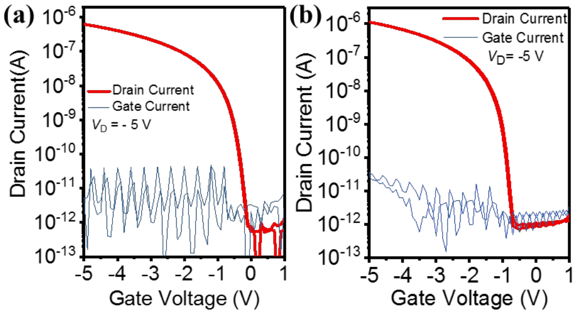

Fig. 2. Transfer characteristics of (a) PVCN based OFET (b) Biopolyamide based OFET

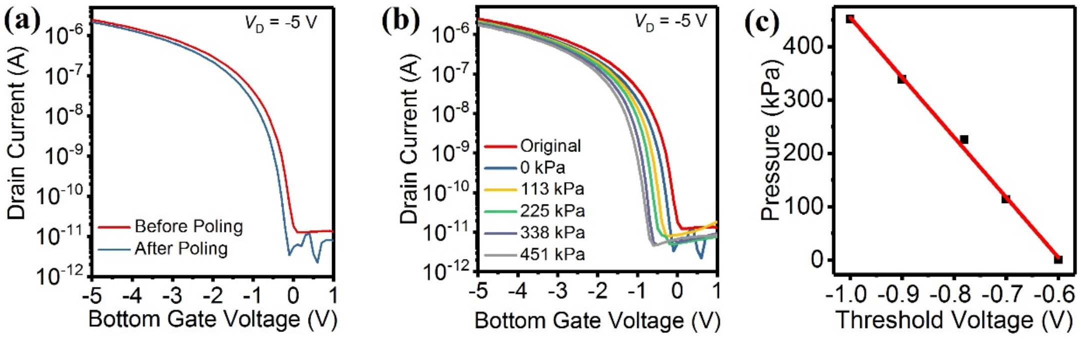

Fig. 3. (a) Threshold shift when polarized P(VDF-TrFE) was placed on the active layer (b) transfer curve shifts corresponding to pressure load (c) linear relationship between pressure and threshold voltage.

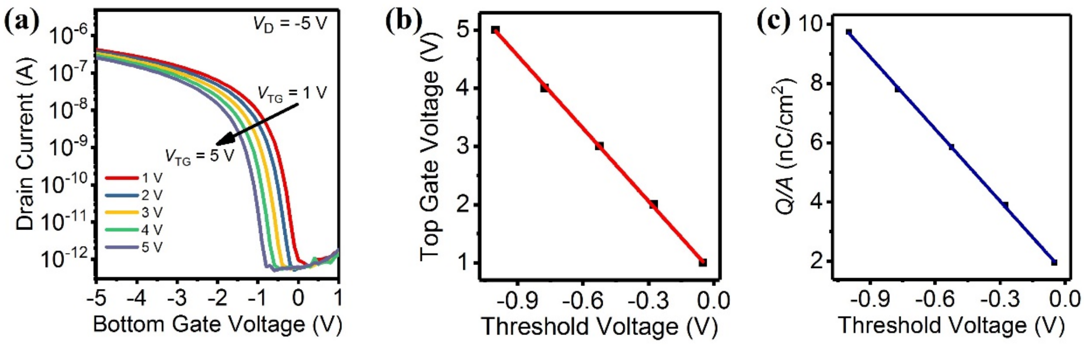

Fig. 4. (a) Threshold shift of the OFET corresponding to top-gate voltage (b) graph of top-gate voltage against threshold voltage (c) graph of charge per unit area against threshold voltage.

Figure 4 (a) shows the shift in transfer characteristics. This shift is consistent with that obtained by the dual-gate organic pressure sensor. In addition, the top-gate voltage relationship to V_{th} is linear (see Fig. 4b). From these results, the amount of charges generated by the piezoelectric P(VDF-TrFE) layer was estimated from the slope of Fig. 4c using equation (1)

In Chapter 5, By a quantitative analysis of results obtained from both the dual-gate pressure sensor and a conventional dual-gate OFET, d_{33} of the sensing layer was estimated. The amount of charges per unit area, Q depleted by the top-gate bias voltage, V_{top} from the equation (2) given below [6]. Fig. 4c shows the relation between Q and the threshold voltage shift.

$$Q = C_{top} V_{top} \ \ldots\ldots\ldots\ (1)$$

Where C_{top} (2.065 nF/cm^2) is the capacitance of the top gate dielectric. In order to determine the piezoelectric constant, d_{33}, equation (2) which states that the quantity of charges, Q, developed by the piezoelectric sensing layer is proportional to force applied, F, with the piezoelectric constant, d_{33}, as the proportionality constant was used [7].

$$Q = d_{33} F \ \ldots\ldots\ldots\ (2)$$

Deducing F from the pressure applied on the 0.87 cm^2 sensing area (A), equation (1) can be modified to this:

$$Q\ V = d_{33} F/A\ V \ \ldots\ldots\ldots\ (3)$$

Both sides of equation (3) were extracted from the slope of Fig. 3b and Fig. 4c which are values c.a. 11.2 × 10^5 Pa/V and 8.1 nC/cm^{2}V, respectively. The absolute value of the piezoelectric constant d_{33} was calculated to be 72 pC/N. d_{33} of the piezoelectric layer was measured directly with a piezoelectric measurement system to be an average of 53 pC/N. These results conclude that the operation mechanism of the dual-gate pressure sensor was due to the piezoelectric behavior of the P(VDF-TrFE) layer used in the device. **In Chapter 6**, the surface charges on the polarized PVDF-TrFE layer required to cause the initial V_{th} shift, with respect to the magnitude of electric field used to polarize the P(VDF-TrFE) layer was quantified using results of a conventional dual-gate OFET. **Chapter 7** summarizes the results achieved in the previous chapters, future research as well as prospective applications of this work.

Part 2: Research Purpose

The main results of this research give a quantitative description of the sensing mechanism of a novel low-voltage dual-gate organic transistor based pressure sensor [5]. Since solution processing methods were used to fabricate these devices, substrates such as polyethylene naphthalate (PEN) and polyethylene terephthalate (PET) could be used to fabricate flexible dual-gate organic pressure sensors the nearest future. In addition, a high-throughput production process can be implemented to commercialize the pressure sensors.

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Part 3: Research Accomplishments

Publications

1. Ogunleye Olamikunle Osinimu, Heisuke Sakai, Yuya Ishii, Hideyuki Murata. “Investigation of the sensing mechanism of dual-gate low-voltage organic transistor based pressure sensor,” Organic Electronics (accepted)

2. Ogunleye Olamikunle Osinimu, *et al.* A degradable biosynthesized polyamide as the gate dielectric for low-operating voltage solution-processable organic field-effect transistors. (Manuscript in preparation)

Conferences

1. Ogunleye Olamikunle Osinimu, Yohei Yoshinaka, Heisuke Sakai, Tatsuo Kaneko, and Hideyuki Murata. “A Biodegradable Biopolymer as Dielectric for Low-Voltage
Solution-Processed Organic Field-Effect Transistors”, International Symposium on Organic

Electronic Molecular Electronics, Saga, Japan. May 31st – June 2nd, 2018.

2. Ogunleye Olamikunle Osinimu, Heisuke Sakai, Yuya Ishii, Hideyuki Murata. “Investigation of the Sensing Mechanism of the Dual-gate Low-voltage Organic Transistor based Pressure Sensor”, International thin-film transistor conference, Okinawa, Japan. Feb. 28th – Mar. 2nd, 2019.

3. Ogunleye Olamikunle Osinimu, Heisuke Sakai, Yuya Ishii, Hideyuki Murata. “Investigation of the Sensing Mechanism of the Dual-gate Low-voltage Organic Transistor for Pressure Sensing by Quantitative Analysis,” JSAP spring meeting, Tokyo, Japan. Mar. 9th – Mar.12th, 2019.

Keywords: Dual-gate pressure sensor; Dual-gate organic field-effect transistor; threshold voltage; P(VDF-TrFE); Piezoelectric constant.

論文審査の結果の要旨

有機発光ダイオード，有機薄膜太陽電池，有機電界効果トランジスタ（OFET）などの有機デバイスは，軽量，柔軟性などの特徴に加えて，溶液プロセスによる大面積・低コスト製造の可能性を有しており，これらを応用したフレキシブル電子デバイスの研究が活発化している．その中でも，温度や圧力といった環境情報を電気信号に変換する有機センサは， IoT社会の基盤となるデバイスとして注目されている．

本論文は，デュアルゲート型OFET（DG-OFET）構造のトップゲート（TG）部分に，圧電性を示す感圧部を用いたDG-OFET型圧力センサに関する研究である．この圧力センサは，本研究室で開発された新規構造であり，高感度かつ低消費電力（駆動電圧５V以下）という優れたデバイス特性を示す．しかしながら，この圧力センサの動作機構については詳らかにされていなかった．本研究では，高性能なDG-OFET型圧力センサの動作機構を明らかにすることを目的として研究に取り組んだ．

TG部分が通常の絶縁体とTG電極の積層構造から構成されるDG-OFETではTG電圧を変化させると閾値電圧（Vth）がシフトする．一方，当該センサの感圧部に圧力を印加するとOFETのVthがシフトする．これら二つのデバイスの構造的類似性から，圧力センサのVthがシフトは感圧層として用いた高分子強誘電体（P[VDF-TrFE]）の圧電効果によってTG部に電位が発生したためと推定された．この仮説を検証するために，DG-OFET型圧力センサと通常のDG-OFETのVthを同じ値だけシフトさせるのに必要なセンサの圧力値とDG-OFETのTG電圧値の関係を定量的に調べた．

具体的には，DG-OFET型圧力センサに0～450 kPa程度の圧力を印加すると，圧力の大きさに比例してVthが線形応答して約0.4 V程度変化した．したがってVthを1 Vシフトするために必要な圧力は，1.12×103 kPaと求まる．一方，DG-OFET のVthはTG電圧に比例して線形応答し，Vthを1 Vシフトさせるために必要な電荷量は8.1 nC/cm2と求まった．すなわち，強誘電体感圧層に1.12×103 kPaの圧力を加えると8.1 nC/cm2

の電荷が発生したことになり，この関係から算出される強誘電体層の圧電定数（d33）は72 pC/Nである．一方，感圧層の高分子強誘電体（P[VDF-TrFE]）膜のd33値を変位測定法によって直接評価したところ53 pC/Nが得られた．このように圧力センサのデバイス特性から推定されたd33値と強誘電体薄膜の物性評価から直接得られた値の間に良好な一致がみられたことから DG-OFET型圧力センサの圧力応答は感圧層の圧電効果によって生じたと結論づけた．

なお，DG-OFET型圧力センサの動作機構の解析に加えて，本研究を実施する過程の中で，OFET型センサの駆動に必要不可欠なOFETの低電圧駆動化がグルコース由来の高分子をゲート絶縁層に用いることで実現できることを見出している．

以上，本論文は，DG-OFET型圧力センサの動作機構について解明したものである．この内容は学術的に貢献するところが大きいと判断し博士（マテリアルサイエンス）の学位論文として十分価値あるものと認めた．