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Laser-Induced Graphene Electrodes for Organic Electrochemical Transistors (OECTs)

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Organic electrochemical transistors (OECTs) have drawn significant interest because of their low cost, biocompatibility, and ease of fabrication, allowing them to be utilized in various applications including flexible displays, electrochemical sensing, and biosensing. Key components of OECTs are the gate, source, and drain electrodes. Herein, OECTs with laser-induced graphene (LIG) electrodes are demonstrated. The electrode patterns for the source, drain, and gate are created by converting the polymer substrate polyimide (PI) into LIG using a scanned laser. The process is simple and inexpensive without complicated chemical synthesis routines or expensive materials such as gold. Patterns can be customized quickly and digitally. The low-cost and porous LIG electrodes with low contact resistance and good electrical stability play an essential role in device performance. The minimum sheet resistance achieved with this laser method for the square patterned electrodes is 7.86 Ω sg⁻¹. The LIG-based OECTs demonstrate good electrical modulation with ON-OFF ratio of 72.80 and high ON current on the order of mA. The LIG-based OECTs exhibit comparable or better performance in comparison with other reports of OECTs on plastic substrates using more complex fabrication methods in terms of OFF current, ON current, transconductance (gm), and contact resistance.

1. Introduction

Organic electronics like organic light-emitting diodes (OLED)^[1] organic solar cells^[2] organic sensors^[3] and organic thinfilm transistors (OTFTs)^[4] have recently attracted a great deal of attention compared to silicon-based devices owing to their low-cost manufacturing, flexibility, and more ecofriendliness. Since their introduction as a form of OTFT, organic electrochemical transistors (OECTs) have seen extensive

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use in monitoring cells^[5] biological sensors^[6] digital logic,^[7] and neuromorphic applications due to their biocompatibility^[8] stretchability^[9] and straightforward fabrication.^[10]

OECT devices contain three electrodes: source, drain, and gate. P-type semiconductor material is deposited between the source and drain without contacting the gate electrode. The main difference compared to organic fieldeffect transistors (OFETs) is the use of an electrolyte instead of the dielectric, which directly contacts the gate electrode. According to the Bernards model, these transistors consist of electronic and ionic circuits.^[11] The gate electrode, which is in contact with the electrolyte, can cause the ions to start moving and diffusing into the semiconductor channel via the applied small gate voltage. Besides this ionic circuit, the electronic circuit can be made by applying a voltage between the source and drain. This drain voltage induces a drain current,

the output current of the OECT. The small value of the gate voltage, the low power requirements, and the high value of the drain current and transconductance mean OECTs are promising for portable applications.^[12,13]

In terms of semiconductor material, poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS) is a promising material that has high conductivity, excellent optical transparency, and biocompatibility. Besides these advantages, PEDOT:PSS-based devices work only in depletion mode since it is naturally in a conductive state.^[14]

The depletion mode is defined as the transistor being in the ON-state without any applied gate voltage owing to PE-DOT being intrinsically doped by PSS (PEDOT⁺: PSS⁻). Applying a positive gate voltage causes the cations (M⁺) in the electrolyte to diffuse into the semiconductor channel, which contains mobile holes (h⁺). This phenomenon decreases the number of holes and the drain current as a result. The semiconductor channel is transformed into an insulating neutral state (PEDOT⁰) from its conductive condition. Conversely, applying a negative gate voltage turns the transistor on. This doping process for the PEDOT:PSS channel is described by Equation (1). The scenario where $V_g = 0V$ is shown on the left-hand side of the equation, while the case where $V_g > 0$ is shown on the right-hand side.

$PEDOT^{+}: PSS^{-} + M^{+} \rightleftharpoons PEDOT^{0} + M^{+}: PSS^{-} + h^{+}$ (1)

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Amine-based molecular de-dopants can be used to achieve accumulation mode operation for PEDOT:PSS based OECTs via changing the doping state in PEDOT:PSS.^[15]

In electronic and optical devices, several materials and methods have been widely used for fabricating electrodes. A promising material, indium tin oxide (ITO), has a high optical transmittance and good conductivity. However, it has drawbacks such as the price of indium, its difficulty to be printed on flexible substrates, and its brittleness which leads to a sharp decrease in the electrical conductivity with bending.^[16] The noble metals gold^[17-19] palladium^[20] and platinum^[21] have long been considered among the best metals for source and drain electrodes due to their high conductivity, stability, and well-matched work function, but a major drawback is their high cost.

In general, not only do the electrode materials themselves have disadvantages such as price, but there are also difficulties with fabricating them. Traditional methods to fabricate electrodes, such as metal evaporation and lithography, are costly, slow, and cannot be customized easily. Photolithography can lead to problems in the fabrication of electrodes on organic semiconductor films since the films can be damaged by the photoresist.^[22] Although Makita et al. transferred metal electrodes to another substrate to prevent the damage from photoresist, there are still several challenges, such as the use of high vacuum and thermal damage during evaporation.^[22] Printed electrodes can overcome some of these limitations; however, this typically still requires multiple steps, synthesis of nanomaterials, and formulation of inks containing nanomaterials, which can be costly.^[23]

To overcome these challenges, graphene-based materials can be an alternative to ITO and evaporated metals due to their advantageous properties such as flexibility and high surface area.^[24] For fabricating graphene-based electrodes, some common methods include chemical vapor deposition (CVD), molecular beam epitaxy (MBE), and chemical synthesis; however, these methods are still challenging in terms of the cost and difficulties during the fabrication process.^[25] Printing techniques are another method to fabricate graphene-based electrodes. It has been reported in several studies that printed graphene electrodes can be used in wearable and flexible electronics.^[26,27] High-resolution graphene patterns can be produced using inkjet, transfer, and gravure printing techniques, whereas screen-printing can create low-resistance thick films in a single pass, which shortens processing times. The challenge of completely eliminating the binder or surfactant elements of the inks from the printed patterns is a challenge. To improve the stability and printability of graphene inks, a binder or surfactant coats the graphene flakes, which limits conductivity.^[28] Alternatively, a simple method has been developed to eliminate these disadvantages of traditional electrode materials and fabrication methods. By using a CO₂ laser, a polymeric substrate can be converted to laser-induced graphene (LIG) in a simple, low-cost way as shown in Figure 1.

Although a conducting carbon network was achieved in 1994 for the first time using an ultraviolet (UV) laser,^[29] Lin et al. ^[30] were the first researchers who demonstrated LIG fabrication using a CO₂ laser in 2014. It has been reported that with the use of a

CO₂ laser, LIG can be produced on different polymeric substrates such as polyimide (PI), polyetherimide (PEI), and polysulfone (PSU). Other substrates, such as wood, potatoes, and paper, were also found to be convertible to LIG, demonstrating the breadth of materials on which LIG can be generated.^[31] The aromatic segments in the polymer precursor structures may play a role in the polymer's transformation into LIG. C–O and C–N bonds are thermally decomposed as a result of fast temperature increases, followed by the formation of ring clusters that form graphitic structures.^[32] Laser patterning of graphene as a one-step fabrication method is very simple and cost-effective, which can be employed in large-scale and mass production of LIG sheets. Scanning the laser beam can easily pattern graphene without requiring complex patterning steps such as photolithography.

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LIG is a type of graphene material produced by the irradiation of a solid polyimide film with a laser, which results in the formation of a network of interconnected multi-layer graphene flakes. Monolayer chemical vapor deposition (CVD) graphene, on the other hand, is grown from a gas-phase precursor on a metal substrate. In terms of sheet resistance, studies have shown that laser-induced graphene has a much lower sheet resistance compared to monolayer CVD graphene. This is due to the fact that laser-induced graphene has a much larger thickness than single or multilayer CVD graphene on the order of micrometers.^[30,33] Within LIG flakes, a high degree of graphitization allows for an efficient transport of charge carriers, but conductivity is limited by transport between flakes. By tailoring the laser settings, it is possible to alter the conductivity and porosity of graphene for a variety of uses. These particular characteristics of LIG make it an appropriate candidate to be used as a conductor in supercapacitors^[34,35] electrochemical sensors^[36] batteries^[37,38] and antibacterial applications.[39]

LIG electrodes have been used in micro-supercapacitors, leading to enhanced capacitance due to the large specific surface area of LIG.^[30] LIG electrodes have been used in mechanical sensors because of the high conductivity and good stretchability. In one report, a wavy LIG strain sensor was fabricated by using a wavy plastic mold as a substrate. It was possible to obtain low hysteresis and high gauge factor, which means it can be used for detecting muscle and joint movement as a wearable flexible strain sensor.^[40] OECTs with LIG electrodes can potentially be integrated into textiles to create smart clothing that can detect environmental conditions such as temperature, humidity, and even the presence of pollutants in the air.^[41] Also, OECTs with LIG electrodes could be used to amplify biological signals, such as those produced by neurons in the brain. By amplifying these signals, researchers can better understand the functions of the brain and develop new treatments for neurological disorders.^[42]

Although several materials have been reported as electrodes in OECTs, to the best of our knowledge, there is only one very recent paper on OECTs with LIG electrodes^[43] that did not use scalable techniques for the semiconductor deposition. Here, we employed inkjet printing for precise deposition of the PEDOT:PSS semiconductor channel and plasma treatment for the gate electrode, leading to improved wetting of the electrolyte and lower OFF current. We demonstrate LIG electrodes with low sheet resistance by varying the laser parameters. This article studies the effect of different laser parameters for each electrode on device properties. We present LIG-based OECTs with high ON current, low contact

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Figure 1. a) Flexible LIG pattern on PI substrate. b) Laser-induced graphene process on PI substrate. c) LIG source and drain electrode shape consisting of lines and squares for contact pads. The different laser pulse overlaps are shown for two configurations: square 100 µm, and line (rectangle) 7.5 µm. d) Schematic of the architecture of LIG-based OECTs. e) Photo of the fabricated OECT device with LIG electrodes. f) LIG-based OECT during the electrical measurement.

resistance, and significant ON-OFF ratio compared to other types of electrodes using a very low-cost, simple, and customizable fabrication process.

2. Experimental Section

2.1. Laser-Induced Graphene Fabrication

The substrate was a flexible sheet of polyimide (PI), which was a readily available material (Kapton, 125 μ m thickness) on top of a glass carrier. A 40 W CO₂ laser cutter (Hydra 16A) from Hyrel 3D (Atlanta, GA) was used to laser scribe 5 mm long lines (vector mode) and 2.5 mm by 2.5 mm square shapes (raster mode) on polymer sheets. The wavelength of the laser beam was 10.6 μ m, the laser spot size was 400 μ m, and the focal distance was 8 mm. The spiral mode was used for fabricating both lines and squares. For all studies, the laser power ranges between 25%

and 40% with a pulse spacing of 1,000 dots per inch (DPI) along the scan direction of the laser, and the scan rate is varied between 300 and 1500 mm min⁻¹. All of the laser studies were conducted in ambient conditions. As shown in Figure 1c, the pulse spacing orthogonal to the laser scan direction was 7.5 µm for source and drain line patterns, while it was 0.1 mm for square contact pads and the gate. It should be noted that the 7.5 µm mentioned in the paper is the lateral pulse spacing between two pulses and not the width of the line pattern, which is limited by the laser spot size and could be improved in the future by changing the laser optics and potentially using a shorter wavelength.

2.2. Device Fabrication

The device architecture is depicted in Figure 1d. The LIG source and drain electrodes were directly patterned in two parallel lines on Kapton tape. Since in this configuration, the electrolyte will be placed on top of the source, drain, and gate electrodes, two 2.5 mm by 2.5 mm LIG squares were scribed as contact pads attached to the one end of the source and drain electrodes for electrical probing without touching the electrolyte. As shown in Figure 1e, the gate electrode square is lasered with the same size as the contact pads next to the source and drain electrodes in a co-planar architecture. Since LIG is a hydrophobic material, air plasma treatment (Super Plasmod, March Instruments Inc., Concord, CA) was done with 50 W power and 40 s duration for the gate electrode to allow the electrolyte to wet the gate electrode.

Regarding the ink preparation for the semiconductor channel, 6 wt.% ethylene glycol (EG, Sigma Aldrich) and 0.1 wt.% dodecyl benzene sulfonic acid (DBSA) were added to commercial 1.3 wt.% PEDOT:PSS aqueous dispersion in H₂O (Clevios PH 1000, Hereaus) to improve the morphology and jettability of the ink respectively. Moreover, PEDOT:PSS films were cross-linked using 1 wt.% (3-glycidyloxypropyl) trimethoxysilane (GOPS) to make them insoluble in aqueous solutions. The semiconductor layer in the transistors is generally improved by all these additions in terms of its hole and ion conductivity. Finally, the ink was filtered to ensure that no agglomerated particles remained. At room temperature, the viscosity of the ink was measured with a RheoSense VISC Portable Viscometer (RheoSense Inc., San Ramon, CA). The printing of the semiconductor ink in a rectangular shape on top of the source, drain, and transistor channel was done with a custom-built piezoelectric drop-on-demand (DOD) inkjet printer. With a LabVIEW program (National Instruments, Austin, TX), any design can be printed by moving the printer stage relative to the printhead. The nozzle (MJ-ATP-01-60-8MX, Microfab Technologies, Inc., Plano, TX) has a 60 µm orifice diameter. Drop spacing for printing a line was 65 µm, while the drop spacing between each line in a rectangle shape was 30 µm. Then, the printed pattern was dried at 120 °C on a hot plate for 20 min. Finally, 40 µl of NaCl solution in water with a concentration of 100 mM was deposited as the electrolyte on top of the source, drain, and gate electrodes using drop casting. NaCl is often used as an electrolyte in organic electrochemical transistors due to its high ionic conductivity, stability, and low cost. Additionally, NaCl maintains a stable gate potential during the operation of the transistor, which helps in achieving good signal amplification and maintaining reliable device performance.

The time required for the fabrication process of the LIG transistor electrodes is only 40 s. The remaining OECT fabrication steps take minutes, with the longest process being the 20 min anneal of the semiconductor. This could be further optimized in the future to maximize manufacturing throughput.

2.3. Characterization

A four-point probe resistivity measuring system (Pro4, Signatone, Gilroy, CA) was used to test the sheet resistance for rectangular LIG patterns, and a probe station with a semiconductor parameter analyzer was used to measure the sheet resistance for line patterns (Keithley 4200A). Sheet resistance was calculated www.advmattechnol.de

using Equation (2), where R_s stands for sheet resistance, R for resistance, L for length, and W for width.

$$R_{S} = R \times \frac{W}{L} \tag{2}$$

Since the gate electrode has a square shape, the Van der Pauw method was used to measure its sheet resistance. The effect of laser parameters on the morphology and porosity of engraved patterns was studied using a field emission scanning electron microscope (FE-SEM) (Thermofisher Quanta 3D). Raman spectra were recorded using a HORIBA Scientific (model: LabRAM ARAMIS) Raman Microscope using 532 nm laser excitation with a laser power of 50 mW. The areas under the peak were calculated using Fityk software. The Raman peaks were fitted with a Voigt function.

X-ray photoelectron spectroscopy (XPS) measurements were conducted using a K-Alpha XPS system (ThermoFisher Scientific, East Grinstead, UK). The monochromatized Al K-Alpha Xray source with a 150 μ m spot size was used, and charge neutralization was achieved using a low-energy electron/ion (Ar) flood source. Survey spectra were acquired at a pass energy of 200 eV to identify detectable surface species, while high-resolution regional scans were performed at a lower pass energy of 50 eV for quantification. The spectra were collected at a 90° angle relative to the surface plane. ThermoFisher's Avantage software was utilized for data processing, including the calculation of surface elemental compositions based on background-subtracted peak areas derived from transmission function-corrected regional spectra.

Optical profilometry (Bruker contour gt-k, Billerica, MA) was used to measure the thickness of the semiconductor channel. A Keithley 4200A semiconductor parameter analyzer was used to measure the transfer and output characteristics, as shown in Figure 1f. For transfer curves, the drain voltage (V_d) was fixed at -1 V, while the gate voltage (V_g) was varied in steps of 0.02 V from +1 to -1 V. The devices were initialized by performing the transfer measurement twice before recording the transfer curves presented here. V_d was swept in steps of 0.005 V from 0 to -1 V for output curves. This procedure was performed for V_g with increments of 0.5 V sweeping from +1 to -1 V.

3. Results And Discussion

Figure 2a,b shows the effect of different laser parameters (power and feed rate) on sheet resistance for lines and squares. All lines drop in sheet resistance first as laser power increases. Increasing the feed rate from 300 to 1500 mm min⁻¹, the sheet resistance increases. The lowest achieved value is 9.48 Ω sq^{-1} at a feed rate of 300 mm min⁻¹. Moreover, by keeping the feed rate parameter constant at 500 mm min⁻¹ and increasing the laser power from 28 to 40%, 10.86 Ω sq⁻¹ is the lowest sheet resistance that can be obtained. When the laser power is raised above 40%, the material starts to be ablated, and both increasing the laser power and lowering the feed rate reduce the chance of forming LIG. In terms of feed rate, the sheet resistance increases by increasing the feed rate from 300 to 1500 mm min⁻¹. The feed rate of 300 mm min⁻¹ and power of 35% result in the lowest sheet resistance of 7.86 Ω sq⁻¹. Same as other reports,^[44,45] it is also shown here that sheet resistance of the square is lower than the line for most of the

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Figure 2. The effect of a) feed rate and b) power on sheet resistance of lines and squares. The feed rate of 1000 mm min⁻¹ and power of 35% for lines are held constant, while for squares the constant feed rate and power are 500 mm min⁻¹ and 35%, respectively. XPS results show the effect of power on c) carbon and d) oxygen and nitrogen contents. e) Raman spectrum of laser-induced graphene fabricated on PI tape with the laser feed rate of 1500 mm min⁻¹ and power of 35%.

power and feed rates, which is due to more pulses overlapping in the square pattern. With larger power, the sheet resistance increases because of LIG ablation.

The XPS results clearly demonstrate the influence of laser power on the carbon, oxygen, and nitrogen contents of the sample. The results support the anticipated relationship between carbon content and resistance, revealing the expected anticorrelation. As shown in Figure 2c, at low power, the carbon percentage increases with increasing laser power, while the oxygen and nitrogen content decreases as shown in Figure 2d. This indicates that at lower laser powers the polymer has not been fully converted to LIG yet. Conversely, as the laser power increases beyond 35%, an opposite trend is observed for carbon and oxygen. Above this limit (>35%), the material begins to undergo oxidation and starts to become ablated due to the higher energy input during the fabrication process.

In Figure 2e, the Raman spectrum is shown for LIG with a feed rate of 1500 mm min⁻¹ and power of 35%. It is possible to identify the presence of graphene due to the existence of graphene's distinctive peaks. The D, G, and 2D peaks appear at wavenumbers of 1350, 1550, and 2700 cm⁻¹, respectively, indicating the presence of multilayer graphene.

These Raman peaks can be used to determine various graphene structural characteristics. The most significant peak for





Figure 3. SEM images of the LIG surface of line shapes with a) 300, b) 500, c) 1000 mm min⁻¹ laser feed rate, and d) 28%, e) 32%, and f) 35% laser power on PI substrate. The power of 35% and feed rate of 1000 mm min⁻¹ are held constant, respectively.

graphene or graphite, commonly known as the graphitic peak, is the G band. The G/D ratio is commonly used to characterize the defects and disorder in graphene. The ratio of the areas under the G/D peaks is 1.23. The disorder in sp² hybridized carbon structure in graphene can be caused by a variety of reasons. Some of the common causes of such disorder in LIG include defects or structural irregularities in the crystal lattice. These defects may include vacancies (missing carbon atoms), substitutional impurities (e.g., nitrogen or boron atoms replacing carbon atoms), or topological defects such as pentagonal and heptagonal rings that disrupt the regular hexagonal pattern of the graphene lattice. Causes of disorder in LIG may include environmental factors such as exposure to high temperatures, radiation, or chemical interactions that can lead to surface contamination or damage to the graphene structure.^[46,47]

Regarding the number of layers created by laser treatment, the 2D/G Raman peak ratio is often used as an indicator of the number of graphene layers present in a sample. A higher ratio typically indicates fewer layers of graphene (i.e., monolayer or few-layer graphene), while a lower ratio typically corresponds to more layers (i.e. multilayer graphene) with a ratio of 0.5 indicating graphitic carbon. By analyzing the Raman spectrum of the resulting LIG with power of 35% and feed rate of 1500 mm min⁻¹, the ratio of areas under 2D/G peaks is 0.62. The 2D peak has only one peak, which means the material is turbostratic graphitic carbon, in contrast to crystalline graphite with regular AB stacking, which contains multiple 2D peaks.^[48,49]

To demonstrate that laser-induced graphene is a hydrophobic material, water contact angle measurements were performed using the sessile drop method, as shown in Figure S1a,b, Supporting Information, before and after plasma treatment. A higher contact angle indicates a more hydrophobic surface. LIG samples exhibited hydrophobic behavior with an average contact angle of $\approx 97^{\circ}$, while after plasma treatment the contact angle was reduced to 35°, demonstrating that plasma treatment was effective in changing the hydrophobic LIG surface to hydrophilic.

To investigate the porosity of LIG, Figure 3 shows the morphology of both line and square shapes of LIG for different laser power and feed rate. As shown in Figure 3a-c, LIG morphology is more porous with lower feed rates of 300 and 500 mm min⁻¹ compared to 1000 mm min⁻¹ with a constant power of 35%. This difference is also observed for different powers where the porosity of LIG with the power of 35% is larger than for powers 32% and 28% with a constant feed rate of 1000 mm min⁻¹. Hence, by decreasing the feed rate and increasing the power, highly porous LIG is achieved until the material is ablated. This effect is much clearer for lines since there is no overlap between adjacent pulses unlike squares affecting the morphology of LIG. On the edges of the lines, the laser energy density is lower than at the center due to the Gaussian beam profile and fewer pulses overlapping, which causes the flat morphology compared to the porous structure at the center of the line because less gas is released at low energy density.^[50] Since the color of the boundary region is quite similar to the LIG line itself, this area is investigated via Raman spectroscopy. As shown in Figure S2b, Supporting Information, no peaks are found in the Raman spectrum, demonstrating that LIG is not formed in the boundary region around the line. By measuring the conductivity of this region, we found that the material in this region is not conductive, which further demonstrates that no LIG is formed in this region. It is worth mentioning that this boundary region around the LIG source and drain lines is not included in the quoted channel length of the devices.

It is crucial to take into account the thickness of the inkjetprinted semiconductor channel in the OECT devices. The thickness of the semiconductor in the transistor channel, between the source and drain electrodes, was measured using optical profilometry, as shown in Figure S3a,b, Supporting Information. The data indicates that the PEDOT:PSS channel is ≈1500 nm thick. The thickness of the semiconductor layer is an important parameter determining OECT performance. For investigating the effect of semiconductor layer thickness, transistors were fabricated with semiconductor thicknesses of one, two, and three inkjet-printed layers. As depicted in Figure S3c,d, Supporting Information, adding more layers of semiconductor material increases both the OFF and ON currents. Both ON and OFF current follow an increasing trend as anticipated, given the physics of the device. In OECTs, ions travel throughout the whole thickness of the semiconductor, in contrast to FETs, where charges create the channel at the interface between the dielectric and semiconductor. As a result, the hole current can flow through a thicker semiconductor layer as the semiconductor channel thickness increases, which results in a larger ON current. All other experiments shown in this work are done by using one layer of the PEDOT: PSS semiconductor layer.

A transfer curve for the LIG-based OECT without plasma treatment of the gate is shown in **Figure 4**a in the saturation regime. Laser power and feed rate parameters were the same for source, drain, and gate electrodes configured at 35% and 500 mm min⁻¹, respectively. As shown in Figure 4a, the magnitude of the drain current reduces as V_g becomes more positive for a constant $V_d = -1V$. The anti-clockwise hysteresis related to slow ion migration in the double-sweep measurements is very small. As shown in Figure 4a, the OFF current is 0.395 mA. The ON current and ON-OFF ratio for the device are 3.62 mA and 9.16, respectively. The device operates in depletion mode. Transconductance, $g_m = \frac{\partial l_d}{\partial V_g}$, of 1.77 mS was achieved.

The output characteristics are presented in Figure 4b. At low

drain voltages, the channel current has a linear relationship with V_d and saturates at higher V_d levels for different V_g values. Many other transistors, like OTFTs with a solid polymer dielectric, require high operating voltage on the order of several tens of volts,^[51] while in OECTs the operating voltage is significantly lower because OECTs use an electrolyte and rely on an electrochemical reaction, which does not require high voltages, unlike the large electric field that needs to be applied to a solid dielectric to turn on and off organic semiconductors.^[52] However, the transistor is not fully turned off, as shown in Figure 4a,b.

One of the main factors in improving the performance of the obtained OECTs, especially in terms of the OFF current and ON–OFF ratio, is the contact of the gate electrode with the electrolyte on top. For this purpose, the gate electrode was first lasered, and then plasma treated with a power of 50 W for 40 s. This was effec-

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tive in making the hydrophobic LIG gate electrode hydrophilic. The better contact with the NaCl electrolyte allows the transistors to have a much lower OFF current. As shown in the transfer curve in Figure 4c, OFF current of 0.035 mA, ON current of 2.56 mA, and ON–OFF ratio of 72.80 was achieved. The transconductance of this device exceeds 2 mS. The threshold voltage was 0.444 mV (Figure 4d).

The effect on the device performance of laser parameters for the LIG gate and the source/drain electrodes is investigated, as shown in Figure 5. The value of g_m does not change significantly with different values of the LIG laser parameters feed rate and power. This data suggests that the OECT performance is robust with regard to these LIG parameters within the window where conductive LIG is created. A similar result is found for the ON and OFF current (see Figures S4 and S5, Supporting Information). This means the OECT performance is robust to variations in the LIG manufacturing process. The standard deviation was used to calculate the error for Figure 5. Five transistors were fabricated for each laser parameter, and their I-V characteristics were measured to calculate the transconductance. The standard deviation was calculated and indicated on the plot at different laser parameters. This error could be due to several sources such as humidity of the environment, oxidation of the semiconductor, and fabrication steps like manual drop-casting of electrolyte.

Based on the transmission-line method (TLM), an OECT can be considered as a channel resistor (R_{CH}) in series with the contact resistance (R_{C}) formulated in Equation (3).

$$R_{tot} = \frac{\partial V_D}{\partial I_D} = R_{CH} + 2R_C \tag{3}$$

For various gate voltages, the total resistance R_{tot} was calculated from the linear regime of the I_d - V_d characteristics and with channel lengths varying from 150 to 510 µm, as shown in Figure 6a. The resistance changes linearly with the channel length, with slope proportional to the resistance in the channel $(R_{\rm CH})$. The lines can then be extrapolated to zero channel length, which gives 2R_C. The contact resistance of one electrode for different gate voltages is shown in Figure 6b, which shows that $R_{\rm C}$ is highly dependent on V_g . While there are not many reports about the contact resistance of OECTs, the R_c value obtained at different gate voltages in this work is low compared to these reports. Paterson and coworkers^[53] investigated electrode modification and its impact on contact resistance in n-type OECTS. Gold electrodes were fabricated using a lift-off process method with OECT channel width of 100 µm. It was demonstrated that modifying the gold electrodes with thiol-based self-assembled monolayers (SAMs), 4-methylbenzenethiol (MBT), and pentafluorobenzenethiol (PFBT), is helpful for reducing the contact resistance. However, the lowest contact resistance for both source and drain electrodes achieved for $V_{\rm g}$ = 0.5V was more than 1000 Ω cm, while in this work, the contact resistance of 46.24 Ω cm is obtained for one electrode at $V_{\rm g} = -0.5$ V. In another report, the gold electrodes were deposited by thermal evaporation and patterned using photolithography. PEDOT:PSS was spin-coated as the semiconductor channel. The reported contact resistance for one electrode at $V_{\rm d} = -0.3$ V and $V_{\rm g} = 0.1$ V was $\approx 200 \ \Omega$ cm. For the same gate and drain voltage here in this work, a contact resistance of 84.3 Ωcm is obtained for one electrode.^[54]

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Figure 4. a) Transfer and b) output curves of LIG-based OECTs without plasma treatment of the gate. c) Transfer curve and d) output characteristic of OECT with a plasma-treated gate electrode. e) Transconductance versus gate voltage for the OECT with plasma-treated gate electrode. (Power of 35% and feed rate of 500 mm min⁻¹ for gate electrode, power of 35% and feed rate of 1000 mm min⁻¹ for source and drain electrodes).

The drain current versus time was monitored to examine the time response of LIG-based OECT. According to Figure 6c, there is a steady state in drain current for the first few seconds when the gate voltage is not applied. The drain current is greatly decreased when a positive gate voltage of +0.5V is supplied, as expected. The rate at which electrolyte ions diffuse into the semiconductor channel determines how long it takes for the drain current to fall. This decrease of the drain current becomes sat-

urated during the period of time after applying a voltage to the gate.

An electrolyte concentration of 100 mM NaCl was chosen for most experiments to ensure comparability with other relevant studies.^[12,55] Additionally, we investigated the influence of electrolyte (NaCl) concentration on the drain current in the LIG OECT. The OECT was immersed in a beaker filled with NaCl electrolyte, and additional electrolyte was gradually added to increase

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Figure 5. Effect of laser parameters of source and drain a,c) power, b,d) feed rate and laser parameters of gate electrode (a) power, (b) feed rate on transconductance. The feed rate of 1000 mm min⁻¹ and power of 35% are held constant, respectively.

the concentration. As shown in Figure 6d, our experimental results reveal a clear correlation between NaCl concentration and the drain current. As anticipated, an increase in NaCl concentration results in a corresponding increase in drain current, indicating enhanced conductivity within the channel. Therefore, the performance of the transistors presented in this study could be improved further by choosing an optimal electrolyte concentration to maximize the ON-current. Human sweat contains a similar NaCl concentration (\approx 130 mM) to the electrolyte used in our experiments, which presents an opportunity to use the LIG OECT for sweat sensing.

For the LIG-based OECTs demonstrated in this article, the source, drain, and gate electrodes can be fabricated in a few seconds, and the fabrication process is considerably simpler than other methods. Moreover, as shown in Table 1, LIG-based devices exhibit better performance in comparison with many of the reports using metal electrodes on plastic substrates in terms of OFF current, ON current, and g_m . The sheet resistance of the laser-induced graphene reported here is 7.86 Ω sq⁻¹, while the

graphene ink for the gate in reference^[56] (see Table 1) has a much higher sheet resistance of $\approx 26 \text{ k}\Omega \text{ sq}^{-1}$, which may limit performance, and the silver ink for the source and drain electrodes has a comparable sheet resistance of $3.0 \Omega \text{ sq}^{-1}$. The resolution of the LIG-based electrode fabrication method is better than the printed results shown in Table 1. Some reports using traditional e-beam evaporation and lift-off exhibit better resolution; however, theyare more complex and costly processes. The carbon-based electrodes fabricated by 3D printing have longer channel length than this work limited by their resolution. The LIG resolution is limited by the laser spot size, which could potentially be improved in the future. Despite of this limitation, LIG-based OECTs exhibit comparable performance to OECTs with metal electrodes in Table 1.

In terms of potential applications, the OECTs with LIG electrodes demonstrated in this work could be used in energy storage applications with devices such as batteries or capacitors, due to their high output current. The LIG-based OECTs could be used for switching in power applications, for example, powering internet of things (IoT) devices. For this purpose, they could be

Electrode material	Electrode fabrication	Substrate	Channel material	Channel fabrication	Gate material	(mn) W	(mm)	l _{on} (mA)	l _{OFF} (mA)	gm (mS)	Voltages (V)	Ref.
Au	E-beam evaporator	S	PEDOT:PSS	Spin coating	PEDOT:PSS-Ag/AgCI	S	01	0.45	0.05	0.98	$V_{\rm d} = -0.6V /$ $V_{\rm g} = 0.6$ to 0V	[z1]
Au	E-beam evaporator	S	PEDOT:PSS	Spin coating	Multilayer graphene (MLG)	I	I	I	I	I	$V_{\rm d} = -0.25V$ / $V_{\rm g} = 1$ to -0.5V	[57]
Au	Lift-off process	Glass	PEDOT:PSS	Spin coating	Au	Ŋ	Ŋ	2.2	I	2.4	$V_{\rm d} = -0.6V / V_{\rm g} = 0.8$ $V_{\rm g} = 0.8$ to 0V	[58]
Au	Lift-off process	S	PEDOT:PSS	Spin coating	SiO ₂	20	20	4	0.5	1.2	$V_{\rm d} = -0.6V /$ $V_{\rm g} = 5 \text{ to}$ 0V	[18]
Au	E-beam evaporator	Poly-ethylen- naphthalate (PEN)	PEDOT:PSS	Spin coating	Nafion/rGo/carbonized silk fabric (CSF)	6000	200	4.2	0.2	I	$V_{\rm d} = -0.1V /$ $V_{\rm g} = 2 \text{ to}$ 0V	[59]
Ag	Inkjet printing	ā	PEDOT:PSS	Spin coating	Graphene	3 000	1000	0. 195	0.14	0.14	$V_{\rm d} = -0.4V / V_{\rm g} = 1.6$ $V_{\rm g} = 1.6$ to 0V	[56]
Ag	Inkjet printing	đ	PEDOT:PSS	Inkjet printing	Ag	3 000	1000	0.46	0.02	1.2	$V_{\rm d} = -0.4 V / V_{\rm g} = 1.6$ $V_{\rm g} = 1.6$ to 0V	[56]
Fluorine doped tin oxide (FTO)	Chemical etching	FTO	PEDOT:PSS	Spin coating	610	I	I	0.25	0.025	0.177	$V_{\rm d} = -0.6V / V_{\rm g} = 2.5$ $V_{\rm g} = 2.5$ to 0V	[60]
Carbon-filled polylactide (c-PLA)	3D printing	Termoplastic co-polyester (TPC)	PEDOT:PSS	Inkjet printing	Ag/AgCI	10000	3000	0.125	0.05	0.25	$V_{\rm d} = -0.6V /$ $V_{\rm g} = 0.6$ to 0V	[[6]]
LIG	Lasering process	ā	PEDOT:PSS	Inkjet printing	LIG	5 000	190	2.564	0.035	2.2	$V_{d} = -1V / V_{g} = 1 \text{ to}$ -1V - 1V	This work

Table 1. Comparison of OECTs characteristics.

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Figure 6. a) The effect of channel length on the total resistance of OECTs with various gate voltages. b) Contact resistance with respect to the gate voltage. (c) Id-t curve for Vg = +0.5 V representing the role of ions in the OECT. d) Effect of electrolyte (NaCl) concentration on the drain current.

combined with LIG-based supercapacitors or batteries that utilize the highly porous morphology of LIG. The fabricated LIGbased OECTs can potentially be used for measuring electrocardiogram (ECG) signals because they allow for high sensitivity and flexibility. The high conductivity of LIG and high transconductance achieved in this work enables the OECT to detect weak signals that are generated by the heart. Furthermore, the flexibility of LIG allows the sensor to conform to the skin, making it more comfortable to wear than traditional ECG sensors. Therefore, LIG-based OECTs could be a suitable option for measuring ECG signals. Also, OECTs with LIG electrodes can potentially be used as biosensors for the detection of proteins, DNA, and other biomolecules. The LIG possesses a large surface area, which makes these OECTs ideal for biosensing applications. The achieved porosity in this work can potentially make LIG-based OECT more suitable for biosensing applications by increasing the surface area available for biomolecule immobilization.

4. Conclusions

In this article, the fabrication of OECTs using LIG electrodes is demonstrated. These LIG electrodes are made on PI substrate by using a CO₂ laser. Increasing the power and decreasing the feed rate leads to highly porous LIG with lower sheet resistance until the material is ablated. The laser process used to create the square-patterned electrodes yields a minimum sheet resistance of 7.86 Ω sq⁻¹. In the reported OECT, a low OFF current was achieved with plasma treatment of the gate electrode. This plasma treatment increases the exposed surface area of LIG to the electrolyte deposited on top. The results show that devices with the plasma-treated gate electrode demonstrate very low OFF current and high ON-OFF ratio of 72.80. The channel length effect was studied in OECTs showing the linear trend with the total resistance of the devices. Low contact resistance is achieved by using the LIG electrodes. The OECT is robust under different laser parameters leading to consistent device performance. The OECTs with LIG electrodes demonstrated in this paper can be used in various applications like biosensing, energy storage devices, and amplifying biological signals due to their high transconductance, large surface area, and high electrical conductivity. It is important to note that these are not the only applications for OECTs with LIG electrodes, as the technology is still new and there is much research to be done to fully explore its potential.

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Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

digital manufacturing, inkjet printing, laser-induced graphene (LIG), organic electrochemical transistors (OECT), PEDOT:PSS, polyimide

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