A solid state aqueous electrolyte-gated field-effect transistor as a low-voltage operation pressure sensitive platform

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Flexible pressure sensor is increasingly impacting a wide variety of novel applications such as wearable health care sensor, *in-vivo* monitoring, and even artificial skin. As a fundamental component, organic field-effect transistors (OFETs) are an active research area due to their inherent advantages related to low-cost solution fabrication processes and compatibility with plastic substrates. During OFET fabrication, it is almost impossible to avoid the water traces in the OSC active layer, especially when ambient solution processing techniques are employed. Water exhibits a strong influence on the electrical performance in OFETs, such as hysteresis and non-ideal transfer characteristics. Here, we show that the presence of water in OSCs also results in pressure-sensitive devices caused by the modification of the water dipoles alignment. This exciting phenomenon has been exploited in a novel OFET, namely hydrogel-based electrolyte-gated organic field-effect transistor (HYGOFET), where a soft water-based hydrogel layer has been employed as dielectric layer. The hydrogel layer plays two major contributions: (1) providing a constant saturated humidity environment and (2) reducing the operation voltage. The HYGOFET exhibits a high electrical performance and relative long-term stability. Importantly, this device also exhibits an excellent pressure response in the low pressure regime (< 10 kPa) working with a very low power-consumption.

1. Introduction

Flexible pressure sensors have been extensively investigated in the past few years because they are critical components for future practical electronic applications, such as for roll-able displays, electronic skin and for the next generation of wearable health care devices.^[1–6] For the majority of these applications, the main requirements are: low production cost, reduced power consumption and high flexibility to allow device integration on curved surfaces and eventually on human skin.^[2] Considering these needs, organic-based electronics constitute an attractive option since organic materials are compatible with low-cost fabrication processes and can be deposited at low temperature and, thus, on plastic supports. Pressure sensitivity in low-pressure regime (< 10 kPa) is essential for these kinds of applications. Up to now, three transduction mechanisms have been mainly used to convert pressure stimuli into electrical signals:^[2] piezoresistivity,^[7–9] piezoelectricity^[10,11] and capacitance changes.^[1,4,5,12]

Organic field-effect transistors (OFETs) have successfully been employed as electrical transduction platforms in pressure sensor devices. OFETs can be used in devices to transmit information from pressure-responsive components^[13–15] or, alternatively, their electrical response to pressure can also be exploited.^[3,6,16] In the latter case, the most explored route has been focused on the capacitance changes that the dielectric layer suffers when it is exposed to pressure due to geometrical changes and their impact on the OFET performance.^[3,6] Despite the extremely encouraging results that have been achieved so far, unfortunately, most of these devices require a high operation voltage (> 40 V) and thus, high power consumption. Only very recently, a suspended organic semiconductor/dielectric/gate device in which a polyelectrolyte layer was used as dielectric has been reported paving the way to a new generation of low voltage organic pressure-sensor devices. The driving mechanisms of these devices was the modification of the contact between the dielectric and the organic semiconductor due to the presence of an air-gap.^{[17][18]}

The commercialization of low-cost organic electronic devices materials is closely linked to high throughput fabrication processes mainly based on roll-to-roll (R2R). Such processes entail the deposition of inks of the organic semiconductor in ambient conditions. Thus, the presence of traces of water in the active thin films, which is known that can affect the device performance,^[19,20] cannot be completely avoided. In this work, we demonstrate that the application of pressure in OFETs where water traces are entrapped leads to changes in the water dipoles orientation affecting the device response. In order to control such response and achieve a low-voltage operation device, a hydrogel, which is a complex 3D hydrophilic polymeric network capable to hold high amounts of water,^[21–23] has been applied as dielectric,^[24–26] here named after as hydrogel-gated organic field-effect transistor (HYGOFET). In these devices the hydrogel provides a constant water reservoir and guarantees the organic semiconductor current modulation in a reduced voltage window (< 0.5 V) due the formation of electrical double layers (EDLs) upon the application of a gate voltage. The flexible HYGOFETs fabricated show high electrical performance and long-term stability and, further, can be turned into pressure sensors, exhibiting high reproducibility and sensitivity in the low-pressure regime (several hundred Pa to 9 kPa). Our results not only describe a simple method to achieve high electrical performance of HYGOFETs but also pave the way to a novel generation of low-consumption pressure sensitive devices with a unique driving mechanism based on the water dipoles alignment within the organic semiconductor.

2. Results and Discussion

Bottom-gate (Si/SiO₂) bottom-contact (Cr/Au) OFETs based on blends of 2,8-difluoro-5,11-bis(triethylsilylethynyl)anthradithiophene:polystyrene (diF-TES-ADT:PS, **Figure 1a**) deposited by bar-assisted meniscus shearing (BAMS),^[27,28] a technique compatible with R2R processes, were prepared according to a previously published protocol.^[29,30] The devices prepared and measured in ambient conditions exhibited a remarkable *p*-type field-effect

response with a mobility of $0.8 \text{ cm}^2/\text{Vs}$ and a threshold voltage (V_{th}) of -1.5 V (Figure 1b and Figure S1, Supporting Information) in the saturation regime. However, due to the processing of the organic semiconductor (OSC) in ambient conditions, the presence of water traces inside the active film is inevitable, which is translated in a visible hysteresis and a nonlinear dependence of $I_{DS}^{1/2}$ versus V_{GS} in the transfer characteristics and a strong V_{GS} -dependent mobility profile of the as-prepared devices (Figure 1b). A similar behavior, including a visible hysteresis and nonlinear dependence of I_{DS} versus V_{GS} was also observed in the linear regime (Figure S2a, Supporting Information). In contrast, after storing the device in a high vacuum chamber ($\sim 10^{-5} \text{ Pa}$) for more than 70 hours in order to “dry” it for removing any trace of water. The OFET characteristics showed more ideal characteristics, with non-appreciable hysteresis and perfect linear dependences of $I_{DS}^{1/2}$ versus V_{GS} both in the saturation (see Figure 1c) and linear regimes (Figure S2b, Supporting Information). Worth noting is the V_{GS} -independent mobility behavior observed in the “dry” device (inset of Figure 1c), which confirmed again the dramatic influence of the water traces on the electrical performance. Remarkably, the hysteresis and the nonlinear dependence of $I_{DS}^{1/2}$ versus V_{GS} reappeared in linear (Figure S2c, Supporting Information) and saturation regime (Figure S3, Supporting Information) after exposing the “dry” sample again in the ambient atmosphere for 48 hours.

Water is known to orientate under the application of an electric field due to its dipolar nature and, further, such orientation can be affected by the application of pressure.^[31–33] Subsequently, we systematically loaded different calibrated weights on top of the channel region of the Si/SiO₂ substrate-based OFET, as illustrated in **Figure 2a**, in order to apply a controlled pressure in the range $0.3\text{-}3 \text{ kPa}$ on the OSC thin film. A polydimethylsiloxane (PDMS) layer was positioned on top of the active area to guarantee the uniform stimulation of the channel region with the calibrated weight. Also, the higher Young module of PDMS compared to the OSC film ensures that not deformation is occurring in the latter. Interestingly, in the as-prepared devices once the pressure was loaded, a negative V_{th} shift and a consequent

I_{DS} decrease was observed followed by a full recovery of the initial transfer characteristics when the pressure was removed (Figure 2a, left). In sharp contrast, the “dry” OFET (*i.e.*, the device stored under high vacuum for 70 hours) did not respond to the loading of all the tested calibrated weights (Figure 2a, right). This type of devices only became again pressure sensitive after being exposed to ambient conditions for 48 hour, as evidenced in Figure S4 (Supporting Information). A similar pressure response was observed in the linear regime (Figure S5, Supporting Information). Although the role of oxygen cannot be unambiguously addressed, the changes in the water dipole orientation within the OSC film induced by the applied pressure can be correlated with the V_{th} shift thus accounting of the reversible electrical response of the device.

This behaviour is in agreement with previous works that suggested that the orientation of water molecules by the applied field affect the local polarization of the OSCs creating states in the band gap.^[34]

According to our experiments, the application of pressure promotes the transient formation of oriented water dipoles that act as traps and shift V_{th} . Despite these encouraging results, the exploitation of such effect in a pressure responsive device was not straightforward due to the difficulty in controlling the amount of water present in the active layers, which could be in turn dependent on the ambient humidity. Further, an addition drawback for applications was the high operation voltage of these transistors which is not desired for realizing low-power consumption devices and might, additionally, promote water dissociation, concomitantly leading to device unstabilities.^[35] In order to circumvent these issues, an alternative device configuration was proposed employing a solid state aqueous dielectric, namely a hydrogel. In this way, the hydrogel can provide a constant saturated water environment to the active OSC layer and, simultaneously, a low operational voltage due to its high capacitance driven by the formation of EDLs.^[29,36] Furthermore, hydrogels are attractive materials for bio-applications since they are biodegradable and possess a degree of flexibility very similar to natural tissue.^[23]

In particular, here agarose was selected as gelling precursor due to its facile manipulation and easy availability.

A transparent water-based agarose gel (1.5 wt.%, Figure 3c) was prepared by means of BAMS by fast pouring a hot agarose solution below the metallic bar followed by the immediate shearing of the solution along the target substrate. Agarose in water jellifies at room temperature due to the formation of a hydrogen-bonded network which contains mobile ions. Afterwards, the agarose film was assembled on top of a “dry” OSC thin film to achieve a top-gate bottom-contact configuration, here defined as hydrogel-gated organic field effect transistor (HYGOFET), as sketched in Figure 2b. The same pressure stimulation protocol was repeated on the HYGOFET device. Interestingly, the electrical response of the HYGOFET towards pressure resembles the trend found in freshly prepared OFETs, *i.e.* a negative V_{th} shift accompanied by a decrease of I_{on} current was observed once the pressure was applied, which in addition displayed full reversibility when the pressure was removed. Noticeably, this device operates in a voltage window below $|0.5|$ V.

Inspired by these results, the HYGOFET architecture was implemented onto a flexible support, *i.e.* Kapton, in order to replace the rigid Si/SiO₂ substrate employed in our first test. The flexible HYGOFET prototype fabricated is shown in **Figure 3a-c** (see experimental section for details of the fabrication procedure). HYGOFET devices based on three different agarose weight concentrations (*i.e.*, 0.75 %, 1.5 % and 3 wt.%) were explored (Figure S6, Supporting Information). The devices with less agarose (0.75 wt.%) exhibited higher current but they responded to a lower pressure range and were less robust. The opposite tendency was found in the 3 wt.% agarose devices. Taking all this into account, we selected the HYGOFET employing a 1.5 wt.% agarose as the best compromise in terms of electrical performance and also due to its higher stability and facile deposition. Thus, our further investigations were focused on these devices.

The thickness of the agarose layer was estimated in the range of 500-1000 μm as confirmed by scanning electron microscopy (SEM, 720 Pa and RH%=100%, Figure S7 Supporting Information.). It should be noticed that due to the nature of the gel, which is mainly composed of water, a shrinking of the film is expected during the SEM experiments because of the restrictions imposed by this technique. The cross-sectional view of the film shows a complex network structure characterized by numerous pores with an average diameter of 20 μm (Figure 3d).^[21]

As shown in **Figure 4a** and **b**, the transfer and output characteristics of the HYGOFET display a typical *p*-type behavior with a negligible hysteresis. The device exhibited average transconductance of 20 μS and 49 μS in the low bias ($V_{DS} = -0.1\text{ V}$) and high bias ($V_{DS} = -0.4\text{ V}$) regime, respectively (Figure S8a, Supporting Information). All the extracted parameters are summarized in Table S1. As evidence in Figure 4c and Figure S8b (Supporting information), the degree of hysteresis of the transfer curves did not show any dependence with the scan rates (136 mV/s, 72 mV/s and 31 mV/s) and the leakage current did not reveal any electrochemical peaks remaining always around 60 nA (Figure S8b, Supporting Information). Furthermore, the electrical behavior of the HYGOFET is regardless of the gate area as displayed in Figure S9 (Supporting Information) and, as previously reported for other ionic electrolytes,^[37] the dielectric film thickness is not affecting the device response since it is the electrical double layer what is determining the device capacitance. The HYGOFET potentiometric sensitivity was found to be of around 100 μV , which is comparable to the values previously reported for the analogous water-gated FET devices (Figure 4d).^[29] The high stability of the devices was confirmed by the shelf-stability measurements, which demonstrated that the devices were still working after two days (Figure S8c and d, Supporting Information). The small loss of current observed was caused by some water evaporation which was not fully avoidable with this configuration.

Subsequently, the device pressure-response was analysed. The first step consisted in performing a first series of measurements recording several I/V to ensure a high electrical stability of the HYGOFET by observing a negligible shift of V_{th} (less than 5 mV) and a perfect overlapping of the transfer curves (Figure S10, Supporting Information). Afterwards, a controlled pressure was applied on top of the HYGOFET by using a series of calibrated weights up to 9 kPa, as shown in **Figure 5a** (see experimental section). It should be noticed that the application of higher pressures was not realised since it deformed the hydrogel irreversibly. The application of a pressure on the HYGOFET induces the response displayed in Figure 5b ($V_{DS} = -0.4$ V). Once a pressure of 2.1 kPa is loaded on the channel area, a negative shift of V_{th} and a consequent decrease of I_{on} are observed. Once the weight is removed, the electrical characteristics recover their initial state. This behavior was also confirmed with a series of increasing weights (Figure 5c), where a gradual V_{th} shift was observed accompanied by a decrease of both I_{on} and I_{off} (see Figure S11a, Supporting Information). The perfect overlap between the initial transfer characteristic and the ones measured after each pressure stimulus confirms the recovery of the agarose dielectric and the absence of any damage at the OSC:PS active layer. A similar behavior was observed in the low bias regime ($V_{DS} = -0.1$ V, Figure S12, Supporting Information). Moreover, it is interesting to observe how the slope of the transfer characteristic presented in Figure 5b and c remains unaltered during the pressure tests, which suggests that the capacitance at the OSC/hydrogel interface is not affected. This was further confirmed by Electrochemical Impedance Measurements (Figure S13, Supporting Information) where it was observed that the phase angle and the impedance modulus are insensitive to any pressure stimuli, which is in agreement with previous works that have proved that the capacitance of ionic gels is not dependent on the gel geometry.^[38] Moreover, the low resistive behaviour in the high frequency region of the Nyquist plot matches with the nature of the hydrogel where traces of salts impurities are unavoidable.

Analysing the transfer curves, it can be observed that all the transfer characteristics exactly overlap when shifting the curves along the V_{GS} axis (Figure S11b, Supporting Information) according to:

$$V_{GS}^{eff} = V_{GS} + V_{offset}, \quad (1)$$

where V_{GS}^{eff} is the effective gate voltage and V_{offset} is the offset voltage that is dependent on the loading pressure.^[39]

The effect of pressure on the HYGOFET electrical response displays a nonlinear behavior as observed in Figure 5d, which was quantified by means of a quadratic fitting in a range of pressure spanning from 300 Pa to 9 kPa according to:

$$V_{offset}(\Delta P) = S' \cdot \Delta P^2 + S \cdot \Delta P + C_0 \quad (2)$$

Here ΔP denotes the loaded pressure.

The sensitivity of our device was approximated to the first order. A sensitivity (S) of -11 ± 2 mV/kPa and -10 ± 2 mV/kPa were estimated in high bias and low bias regime, respectively (Figure 5d and Figure S12d, Supporting Information), although it should be noticed that a lower variability is found applying pressures below 2 kPa. Additionally, the device displays an amplification ranging from 7 to 16 at different V_{GS} as reported in Figure S14, Supporting Information. These data were averaged among 6 devices, whose statistical results are listed in Table S2 (Supporting Information). Again, it is key to highlight the low operation voltage of these pressure sensitive devices. It should also be highlighted that the pressure sensitivity of the HYGOFET is similar to the one found for the standard OFET devices, elucidating that this behaviour is mainly dominated by the water present in the organic semiconductor and not from the hydrogel.

The HYGOFET was also tested under continuous operation as depicted in Figure 5e ($V_{DS} = -0.1$ V and $V_{GS} = -0.4$ V). During the measurements, three different pressures were applied which induced a decrease of the absolute I_{DS} , which was reproducible and proportional to the

applied pressure. The time response of the HYGOFET towards pressure was estimated to be ~ 2 s from the exponential fit of I_{DS} (*i.e.* $\Delta I_{DS} = e^{\pm(V/\tau)}$, see Figure S15 Supporting Information for a detailed view).^[29]

Several groups have correlated that the compression or tensile deformation of OSCs can lead to morphological changes at the micro-scale that result in an increase or decrease of I_{DS} current, respectively.^[16,40] In order to rule out possible effects coming from the mechanical deformation of the OSC induced by the application of pressure, we performed also measurements applying the weights on the back side of the devices (*i.e.*, tensile deformation). However, we observed a negative V_{offset} shift by applying indifferently a tensile or compression strength (Fig. 5b and Fig. S16, Supporting Information). This result discards the possibility that the OSC deformation is playing a major role in the device response.

3. Conclusion

In conclusion, a solid state aqueous electrolyte-gated field-effect transistor has been fabricated employing an agarose hydrogel as dielectric layer (*i.e.*, HYGOFET). This water based gel enables the fabrication of a flexible and compact device and, further, provides a high capacitance dielectric and maintains a constant saturated humidity environment. The HYGOFET exhibited high electrical performance and relative long-term stability due to a homemade PDMS encapsulation structure. The electrical characteristics of this device respond to the application of low-pressures (in the range of several hundred Pa to 9 kPa) revealing a high reproducibility and sensitivity and, importantly, operating below 0.5 V. The origin of this pressure-induced response has been attributed to the changes in the orientation of the dipole of the water molecules which are entrapped in the OSC thin film. A reduction of the total thickness of the platform and an efficient encapsulation might provide more robust devices closer to real prototypes. Thus, although further device optimizations are required for practical applications, this proof-of concept device demonstrates the feasibility of using HYGOFETs as an excellent

platform to develop portable biocompatible, biodegradable and low-consumption pressure sensors taking advantage for the first time of the influence of the water dipoles alignment on the OSC electrical response.

Experimental Section

Materials: Agarose powder (Type I-B, low EEO), polystyrene ($M_w = 10\,000\text{ g mol}^{-1}$), chlorobenzene (CB), and 2,3,4,5,6-pentafluorothiophenol (PFBT) were purchased from Sigma-Aldrich, and 2,8-difluoro-5,11-bis(triethylsilylethynyl)anthradithiophene (diF-TES-ADT, purity > 99%) was purchased from Lumtec. All these commercial materials were used as received. Polydimethylsiloxane (PDMS, Sylgard 184) was obtained from Dow Corning Corporation and prepared following the protocol at the web page: www.dowcorning.com. Kapton[®] polyimide film (300 HN, 75 μm thick) and silver conductive paint (RS 186-3593) were received from DuPontTM and RS Components Pty Ltd, respectively.

Solution Preparation: The diF-TES-ADT:PS blend solution was prepared according to a previously published protocol.^[29,30] The agarose solutions with various concentrations (0.75 wt.%, 1.5 wt.% and 3 wt.%) were prepared by dissolving the agarose powder into MilliQ water (resistivity: at 25 °C 18.2 $\text{M}\Omega\cdot\text{cm}$) under continuous stirring (800 rpm) at 150°C for 5-10 minutes. Once the agarose solution became transparent, it was immediately used for film preparation.

Film deposition and characterization: For the final device fabrication two kinds of substrates (Si/SiO₂ and Kapton) were employed in this work to deposit the OSC:PS blend and agarose gel. In the first case, Si/SiO₂ substrate or Kapton support (1.5×2 cm²) source/drain (*S/D*) electrodes were patterned by photolithography (MicroWriter^{ML} Laser Lithography System) and afterwards Cr/Au (5 nm/ 40 nm) was thermally evaporated (Evaporation System Auto 360 from BOC Edwards). The channel width (*W*) and length (*L*) were 20700 μm and 30 μm , respectively, with a fixed geometrical ratio $W/L = 690$. This substrate was subsequently coated with diF-TES-ADT:PS blend following the procedure reported previously.^[28,29] The second Kapton foil

(3.5×3.5 cm²) was patterned with a gold electrode ($W = 0.25$ cm, $L=3$ cm), *i.e.* the top gate electrode, and used as support for agarose film deposition. The top gate electrode was patterned according to the aforementioned description.

The agarose gel film was deposited on the second Kapton support on top of the metal gate electrode. Gel films were realized by means of Bar-Assisted Meniscus Shearing (BAMS) using a speed of 5 cm/s and a fixed plate temperature of 30 °C since the gelling temperature of normal agarose is around 35 °C.^[41] During gel deposition, the tip of the micropipette should be kept in the hot agarose solution (150 °C) for 30 s to avoid solution gelling inside the plastic pipette. The agarose film was subsequently shaped into a 0.6×0.6 cm² square. The thickness and morphological properties of the prepared agarose gel film (1.5 wt.%) was characterized by scanning electron microscopy in a humid environment without adding a metallisation layer (Quanta 650 FEG).

HYGOFET device assembly: The two parts, namely diF-TES-ADT:PS-coated Kapton support and the one coated by the agarose gel, were assembled together as depicted in Figure 2. Silver conductive paint was used to connect the *S/D* electrodes to external wires. The prepared HYGOFET was sandwiched into two pieces of thin PDMS films (~ 1 mm) to ensure the sealing of the device and to avoid water evaporation. A hole in the center of the PDMS film allows to maintain a fixed area ($A = 0.36$ cm²) during electrical measurements with pressure stimulation and to circumscribe the stimuli on the active area of the device.

Electrical measurements: The diF-TES-ADT:PS films were characterised employing a standard OFET configuration with Bottom Gate/Bottom Contact by using a heavily doped *n*-type silicon wafer featuring thermal SiO₂ (200 nm thick, $C = 17.26$ nF/cm²) as substrate. The HYGOFETs were electrically characterized under ambient conditions using an Agilent 5100A SMU interfaced with the Easy Expert software. The *I-V* transfer characteristics were recorded at a scan rate of 72 mV/s unless otherwise specified. Regarding the hysteresis *vs.* scan rate test, two scan rates (31 mV/s and 136 mV/s) were used with the same V_{GS} window (Fig. 4c). The

potentiometric sensitivity and shelf stability were recorded following the procedure already published.^[29] Prior to all pressure tests, several *I-V* transfer characteristics were recorded to assess the electrical stability of each HYGOFET and once a stable condition was reached, the first *I-V* curve was defined as reference. The transconductance, defined as $g_m = \Delta I_D / \Delta V_G$, was selected to describe the gate voltage ΔV_G modulation of the HYGOFET current at two different I_{DS} potential, namely $V_{DS} = -0.1$ V and $V_{DS} = -0.4$ V.^[42] The *I-V* transfer characteristics in the pressure response test were recorded by placing calibrated weights on top of the interdigitated. For the shelf stability test, we recorded the electrical performance of the same device once per day. The electrochemical impedance spectra (EIS) were performed with Novocontrol Alpha-AN impedance analyzer equipped with POT/GAL 30V/2A electrochemical interface. These measurements were carried out by sandwiching an agarose gel film (1.5 wt.%) between two Au plates. EIS response was recorded by keeping a DC voltage of 0 V and superimposing an AC oscillation of 5 mV; the frequency was varied from 10^5 Hz to 10^{-1} Hz.

Pressure stimulation: The electrical response of the device due to the application of pressure stimuli was recorded by gently placing different calibrated weights on top of the device. The applied pressure was calculated according to the following relation: $P = \frac{m \times g}{A}$, where m is the mass of the calibrated weight, g is the gravity acceleration, and A is the area of the calibrated weight.

Supporting Information

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

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References

- [1] S. C. B. Mannsfeld, B. C. K. Tee, R. M. Stoltenberg, C. V. H. H. Chen, S. Barman, B. V. O. Muir, A. N. Sokolov, C. Reese, Z. Bao, *Nat. Mater.* **2010**, 9, 859.
- [2] Y. Zang, F. Zhang, C. A. Di, D. Zhu, *Mater. Horizons* **2015**, 2, 140.
- [3] G. Schwartz, B. C. K. Tee, J. Mei, A. L. Appleton, D. H. Kim, H. Wang, Z. Bao, *Nat. Commun.* **2013**, 4, 1859.
- [4] B. C. K. Tee, A. Chortos, R. R. Dunn, G. Schwartz, E. Eason, Z. Bao, *Adv. Funct. Mater.* **2014**, 24, 5427.
- [5] S. Kang, J. Lee, S. Lee, S. G. Kim, J. K. Kim, H. Algadi, S. Al-Sayari, D. E. Kim, D. E. Kim, T. Lee, *Adv. Electron. Mater.* **2016**, 2, 1600356.
- [6] Y. Zang, F. Zhang, D. Huang, X. Gao, C. A. Di, D. Zhu, *Nat. Commun.* **2015**, 6, 6269.
- [7] C. L. Choong, M. B. Shim, B. S. Lee, S. Jeon, D. S. Ko, T. H. Kang, J. Bae, S. H. Lee, K. E. Byun, J. Im, Y. J. Jeong, C. E. Park, J. J. Park, U. I. Chung, *Adv. Mater.* **2014**, 26, 3451.
- [8] L. Pan, A. Chortos, G. Yu, Y. Wang, S. Isaacson, R. Allen, Y. Shi, R. Dauskardt, Z. Bao, *Nat. Commun.* **2014**, 5, 3002.
- [9] Z. Wang, S. Guo, H. Li, B. Wang, Y. Sun, Z. Xu, X. Chen, K. Wu, X. Zhang, F. Xing, L. Li, W. Hu, *Adv. Mater.* **2019**, 31, 1805630.
- [10] J. A. R. Canan Dagdeviren, Pauline Joe, Ozlem L. Tuzman, Kwi-Il Park, Keon Jae Lee, Yan Shi, Yonggang Huang, *Extrem. Mech. Lett.* **2016**, 9, 269.
- [11] L. Persano, C. Dagdeviren, Y. Su, Y. Zhang, S. Girardo, D. Pisignano, Y. Huang, J. A. Rogers, *Nat. Commun.* **2013**, 4, 1633.
- [12] M. J. Pereira, M. Matta, L. Hirsch, I. Dufour, A. Briseno, S. M. Gali, Y. Olivier, L. Muccioli, A. Crosby, C. Ayela, G. Wantz, *ACS Appl. Mater. Interfaces* **2018**, 10, 41570.
- [13] T. Someya, T. Sekitani, S. Iba, Y. Kato, H. Kawaguchi, T. Sakurai, *Proc. Natl. Acad. Sci.* **2004**, 101, 9966.

- [14] F. A. Viola, A. Spanu, P. C. Ricci, A. Bonfiglio, P. Cosseddu, *Sci. Rep.* **2018**, 8, 8073.
- [15] R. A. Nawrocki, N. Matsuhisa, T. Yokota, T. Someya, *Adv. Electron. Mater.* **2016**, 2, 1500452.
- [16] I. Manunza, A. Bonfiglio, *Biosens. Bioelectron.* **2007**, 22, 2775.
- [17] Z. Liu, Z. Yin, J. Wang, Q. Zheng, *Adv. Funct. Mater.* **2018**, 29, 1806092.
- [18] Z. Yin, M. J. Yin, Z. Liu, Y. Zhang, A. P. Zhang, Q. Zheng, *Adv. Sci.* **2018**, 5, 1701041.
- [19] G. Chen, N. Matsuhisa, Z. Liu, D. Qi, P. Cai, Y. Jiang, C. Wan, Y. Cui, W. R. Leow, Z. Liu, S. Gong, K. Q. Zhang, Y. Cheng, X. Chen, *Adv. Mater.* **2018**, 30, 1800129.
- [20] P. A. Bobbert, A. Sharma, S. G. J. Mathijssen, M. Kemerink, D. M. De Leeuw, *Adv. Mater.* **2012**, 24, 1146.
- [21] P. Zucca, R. Fernandez-Lafuente, E. Sanjust, *Molecules* **2016**, 21, 1577.
- [22] E. Mercey, P. Obeïd, D. Glaise, M. L. Calvo-Muñoz, C. Guguen-Guillouzo, B. Fouqué, *Biomaterials* **2010**, 31, 3156.
- [23] D. Buenger, F. Topuz, J. Groll, *Prog. Polym. Sci.* **2012**, 37, 1678.
- [24] Y. J. Jo, K. Kwon, Z. U. Khan, X. Crispin, T. Kim, *ACS Appl. Mater. Interfaces* **2018**, 10, 39083.
- [25] M. Berto, C. Diacci, L. Theuer, M. Di Lauro, D. T. Simon, M. Berggren, F. Biscarini, V. Beni, C. A. Bortolotti, *Flex. Print. Electron.* **2018**, 3, 024001.
- [26] I. Cunha, R. Barras, P. Grey, D. Gaspar, E. Fortunato, R. Martins, L. Pereira, *Adv. Funct. Mater.* **2017**, 27, 1606755.
- [27] I. Temiño, F. G. Del Pozo, M. R. Ajayakumar, S. Galindo, J. Puigdollers, M. Mas-Torrent, *Adv. Mater. Technol.* **2016**, 1, 1600090.
- [28] F. G. Del Pozo, S. Fabiano, R. Pfattner, S. Georgakopoulos, S. Galindo, X. Liu, S. Braun, M. Fahlman, J. Veciana, C. Rovira, X. Crispin, M. Berggren, M. Mas-Torrent, *Adv. Funct. Mater.* **2016**, 26, 2379.

- [29] Q. Zhang, F. Leonardi, S. Casalini, I. Temiño, M. Mas-Torrent, *Sci. Rep.* **2016**, *6*, 39623.
- [30] Q. Zhang, F. Leonardi, S. Casalini, M. Mas-Torrent, *Adv. Funct. Mater.* **2017**, 1703899.
- [31] G. Chopra, M. Levitt, *Proc. Natl. Acad. Sci.* **2011**, *108*, 14455.
- [32] Y. He, G. Sun, K. Koga, L. Xu, *Sci. Rep.* **2014**, *4*, 6596.
- [33] B. R. H. de Aquino, H. Ghorbanfekr-Kalashami, M. Neek-Amal, F. M. Peeters, *Phys. Rev. B* **2018**, *97*, 144111.
- [34] K. P. Pernstich, D. Oberhoff, C. Goldmann, B. Batlogg, *Appl. Phys. Lett.* **2006**, *89*, 213509.
- [35] M. Nikolka, I. Nasrallah, B. Rose, M. K. Ravva, K. Broch, A. Sadhanala, D. Harkin, J. Charmet, M. Hurhangee, A. Brown, S. Illig, P. Too, J. Jongman, I. McCulloch, J. L. Bredas, H. Sirringhaus, *Nat. Mater.* **2017**, *16*, 356.
- [36] F. Leonardi, S. Casalini, Q. Zhang, S. Galindo, D. Gutiérrez, M. Mas-torrent, *Adv. Mater.* **2016**, *28*, 10311.
- [37] S. P. White, K. D. Dorfman, C. D. Frisbie, *J. Phys. Chem. C* **2016**, *120*, 108.
- [38] K. H. Lee, S. Zhang, T. P. Lodge, C. D. Frisbie, *J. Phys. Chem. B* **2011**, *115*, 3315.
- [39] D. A. Bernards, D. J. MacAya, M. Nikolou, J. A. Defranco, S. Takamatsu, G. G. Malliaras, *J. Mater. Chem.* **2008**, *18*, 116.
- [40] S. Lai, I. Temiño, T. Cramer, F. G. del Pozo, B. Fraboni, P. Cosseddu, A. Bonfiglio, M. Mas-Torrent, *Adv. Electron. Mater.* **2017**, *4*, 1700271.
- [41] V. Normand, D. L. Lootens, E. Amici, K. P. Plucknett, P. Aymard, *Biomacromolecules* **2000**, *1*, 730.
- [42] D. Khodagholy, J. Rivnay, M. Sessolo, M. Gurfinkel, P. Leleux, L. H. Jimison, E. Stavrinidou, T. Herve, S. Sanaur, R. M. Owens, G. G. Malliaras, *Nat. Commun.* **2013**, *4*, 2133.

Figure 1: a) Molecular structures of diF-TES-ADT and PS. Transfer characteristics of the same diF-TES-ADT:PS OFET device recorded as prepared in ambient conditions (b) and in the “dry” state (c) in saturation regime ($V_{DS} = -15$ V). The “dry” process was carried out by storing the OFET device in a high vacuum chamber ($\sim 10^{-5}$ Pa) for 70 hours. The inserts correspond to the devices mobility profiles. In the first case, the measurement was carried out in the atmosphere environment and in the second case, the measurement was tested in the glove box. The measurements were performed in a bottom gate/bottom contact configuration by using a Si/SiO₂ substrate (200 nm thick, $C = 17.26$ nF/cm²) patterned with interdigitated gold electrodes.

Figure 2: Electrical characteristics of the pressure-induced response performed on Si/SiO₂-based OFETs and on HYGOFETs fabricated on Si/SiO₂ substrates. (a) Schematic diagram of the application of weights on the OFETs (middle). Transfer characteristics recorded in saturation regime ($V_{DS} = -15$ V) for the as-prepared devices (left) and “dry” devices (right) in the presence and absence of the weights. (b) Schematic diagram of the HYGOFET on a Si/SiO₂ substrate subjected to pressure (left) and transfer characteristics of the HYGOFET recorded at $V_{DS} = -0.4$ V when a weight is applied and in the relaxed state (right).

Figure 3: (a) Schematic diagram and (b) a photograph of the flexible HYGOFET. (c) Photograph of the self-standing agarose film (1.5 wt.%) and chemical structure of agarose. (d) SEM image of a cross-section of the 1.5 wt.% agarose film.

Figure 4: Electrical characteristics of the HYGOFET based on 1.5 wt.% agarose gel. (a) Transfer curves at $V_{DS} = -0.1$ V and $V_{DS} = -0.4$ V. (b) Output curves at different gate-source voltages. (c) Transfer characteristics recorded at three different scan rates (136 mV/s, 72 mV/s and 31 mV/s) at $V_{DS} = -0.4$ V. All the solid lines represent the forward sweep and the dashed lines are backward sweeps. (d) I - t plots corresponding to a 100 μV step potential.

Figure 5: Schematic diagram of the pressure test and electrical characteristics of the HYGOFET fabricated on Kapton. (a) Schematic diagram of the pressure setup employed for the measurements. Transfer characteristics ($V_{DS} = -0.4$ V) recorded at (b) constant pressure and (c) by varying the calibrate weights. The inset in b) shows the reversibility of the system after applying and releasing a pressure of 2080 Pa. In c) The pressure is loaded from 300 Pa to 9 kPa in ascending order on a contact area (5 mm^2). All the dashed lines represent the electrical response when no pressure is applied. (d) Plot of V_{offset} versus pressure. (e) $I-t$ plot of the device operated with loaded pressure and relaxed at constant $V_{GS} = -0.4$ V and $V_{DS} = -0.1$ V. The base current shows a drift during the first hour of operation but then it stabilizes to a constant value. The red and blue lines stand for the exponential fits related to the speed response speed towards pressure (see Figure S15 Supporting Information for a detailed view).