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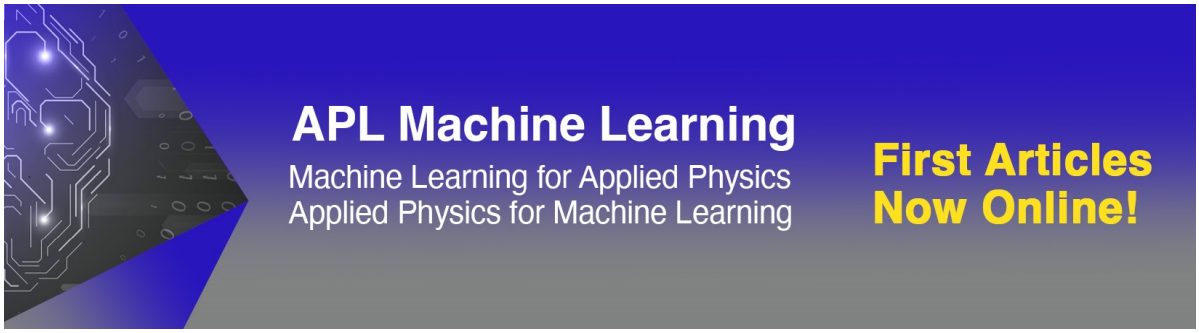

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Solvent-induced changes in PEDOT:PSS films for organic electrochemical transistors

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Organic electrochemical transistors based on the conducting polymer poly(3,4-ethylenedioxythiophene) doped with poly(styrenesulfonate) (PEDOT:PSS) are of interest for several bioelectronic applications. In this letter, we investigate the changes induced by immersion of PEDOT:PSS films, processed by spin coating from different mixtures, in water and other solvents of different polarities. We found that the film thickness decreases upon immersion in polar solvents, while the electrical conductivity remains unchanged. The decrease in film thickness is minimized via the addition of a cross-linking agent to the mixture used for the spin coating of the films. © *2014 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution 3.0 Unported License.* [http://dx.doi.org/10.1063/1.4905154]

Organic electronic devices have attracted particular attention in recent decades because they are easy to process over large areas, exhibit electronic properties that can be tuned via chemical synthesis, and are compatible with flexible and low-weight substrates.[1,2] Besides established applications such as organic light-emitting diodes, organic solar cells, and organic field-effect transistors,[3,4] the past few years have seen the rise of organic bioelectronics, which exploits the coupling of devices based on conducting polymers with biological systems.[5] Among organic bioelectronic devices, organic electrochemical transistors (OECTs) are investigated because they can be operated at low voltages (below 1 V) and are able to act as ion to electron converters.[6–11] OECTs consist of source and drain electrodes and a channel containing a conducting polymer in ionic contact with a gate electrode via an electrolyte solution. The gate voltage modulates the current flowing in the channel between the source and drain electrodes.[12,13] OECTs have been used as sensors for chemical and biological species, such as hydrogen peroxide,[14] glucose,[15] neurotransmitters,[16] chloride ions,[11] DNA,[17] as well as to establish controlled cell gradients on conducting polymer surfaces,[18] monitor tissue integrity,[19] and record *in vivo* brain activity.[20] They have been integrated with microfluidic channels for lab-on-a-chip applications,[21] and are also the fundamental unit of other newly emerging devices, such as ion transistors and organic electronic ion pumps.[9,22]

The majority of OECTs are currently based on the conducting polymer poly(3,4-ethylenedioxythiophene) doped with polystyrene sulfonate (PEDOT:PSS). PEDOT:PSS is a heavily doped p-type semiconductor, where holes on the $PEDOT^+$ chains are compensated by sulfonate anions on the PSS^- chains (dopant).[23] The application of a positive gate bias causes cation inclusion into the PEDOT:PSS channel.[12] These cations compensate the sulfonate anions in the film and de-dope $PEDOT^+$, thereby decreasing the source-drain current. PEDOT:PSS films are typically obtained by spin coating from commercially available aqueous suspensions, which need to be mixed with other chemical compounds to increase film electrical conductivity, which in films processed from pristine PEDOT:PSS suspensions is typically as low as 1 S/cm.[8,15,24] Mixing with other chemical compounds is also necessary to facilitate film processing and to improve film mechanical stability. The compounds employed to increase film conductivity are often named *secondary dopants*. However, as

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there is no evidence that they change the doping level of PEDOT,[25] it is more appropriate to name them *conductivity enhancement agents*.[26] The conductivity increase upon addition of conductivity enhancement agents is likely due to the fact that they alter the film morphology during drying, leading to lower energy barrier for charge carrier transport between individual PEDOT:PSS clusters.[26] Despite the widespread use of PEDOT:PSS in organic electronics, little attention has been so far dedicated to the behaviour of PEDOT:PSS films in water, which is a universal solvent for bioelectronic applications. In this letter, we investigate the variation of the thickness, chemical composition, and electrical properties of PEDOT:PSS films after immersion in water and in other solvents with different polarities. These studies are of paramount importance for the development of devices working at the interface with biological systems and living tissues, where the release of substances needs to be strictly controlled.

PEDOT:PSS films were deposited on glass substrates by spin coating from mixtures containing all or part of the following components: a PEDOT:PSS aqueous suspension (Clevios™ PH1000, Haraeus Electronic Materials), the surfactant dodecyl benzene sulfonic acid (DBSA), which facilitates film processing, one conductivity enhancing agent among glycerol, sorbitol, ethylene glycol (EG), and dimethyl sulfoxide (DMSO), to increase film electrical conductivity, and the cross-linking agent 3-glycidoxypropyltrimethoxysilane (GOPS), to improve film mechanical stability and adhesion to the substrate.[27,28] The Clevios PH1000 PEDOT:PSS aqueous suspension has a PEDOT:PSS content of 1.1 w/w %, with a PEDOT to PSS ratio of 1:2.5 (i.e., ~0.3 w/w % PEDOT and ~0.8 w/w % PSS).[26] Therefore, the amounts of conductivity enhancement agents added to the suspension can, as it is the case for the range of ~2 w/w % and 40 w/w % investigated in this work, significantly exceed the initial amount of PEDOT:PSS. After spin coating, the films were baked at 140 °C for 1 h. The film thickness was measured with a Dektak 150 profilometer. The film electrical conductivity was obtained by four point probe measurements according to the model developed by Smits.[29,51]Additional experimental details are available in the supplementary material.[30]

To gain insight into the effect of the various additives, we first investigated films obtained from a mixture of Clevios PH1000 and DBSA. Although DBSA is widely used as an additive (surfactant) to facilitate film processing from PEDOT:PSS suspensions,[12,20,23] its effect on film conductivity has not been investigated yet. By measuring the electrical conductivity of PEDOT:PSS films processed from mixture with different DBSA contents, we found that DBSA significantly affects film conductivity (inset of Fig. 1). The highest conductivity (~500 S/cm) is observed for a mixture containing

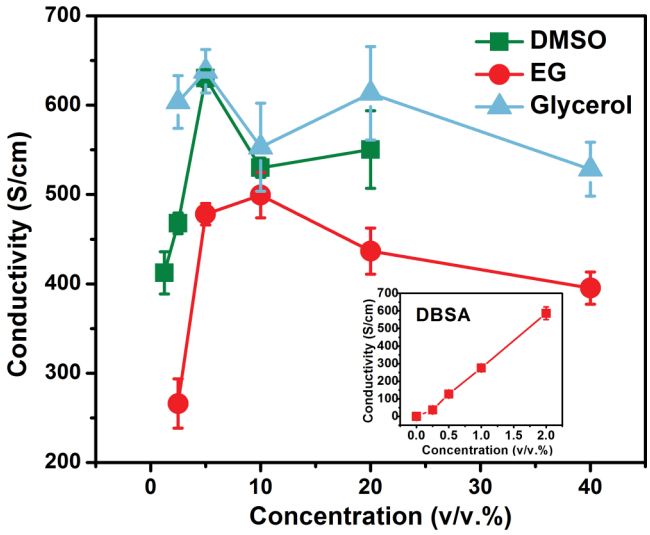

FIG. 1. Electrical conductivity of PEDOT:PSS films processed from mixtures containing Clevios PH1000, 0.5 v/v % DBSA, and different v/v % concentrations of conductivity enhancing agents (DMSO, EG, glycerol). The inset shows the electrical conductivity variation versus increasing DBSA concentrations in absence of conductivity enhancing agents. The films thickness is about 150 nm. The error bars correspond to the standard deviations of three samples.

2 v/v % DBSA. However, we found that concentrations of DBSA higher than ca. 0.5 v/v % induce a phase separation in the mixture, which makes processing by spin coating more difficult and results in a poor film quality. Therefore, for the following studies, we kept the DBSA concentration at 0.5 v/v % and used conductivity enhancing agents, such as glycerol, sorbitol, EG, and DMSO, to achieve high film conductivity. Although conductivity enhancing agents have been used for decades for the processing of PEDOT:PSS films, their role is still under debate. Kim *et al.* proposed that these compounds could induce screening effects, thus reducing the coulombic interactions between $PEDOT^+$ and PSS^- and therefore enhancing the charge carrier hopping rate and conductivity in the PEDOT:PSS films.[31] Inganas *et al.* considered conductivity enhancing agents to act as plasticizers, which would aid the reorientation of the PEDOT:PSS chains during the baking process to form more connection pathways between $PEDOT^+$ chains.[32] Ouyang *et al.* attributed the conductivity enhancement to an effect on the conformation of the $PEDOT^+$ chains.[33] Leo *et al.*[34] and Jonsson *et al.*[35] assumed that removal of insulating PSS^- is the prime reason for the conductivity improvement after the introduction of conductivity enhancing agents. It has been reported recently that the conductivity of PEDOT:PSS films can be increased up to about 4000 S/cm, a value comparable to that of indium tin oxide, via a post-treatment with sulphuric acid.[36] However, sulphuric acid is a strong and corrosive acid, which limits its application in bioelectronics.

Fig. 1 shows the effect of the conductivity enhancing agents EG, DMSO, and glycerol (data for sorbitol are shown in Fig. S1 in the supplementary material[30]) on the conductivity of films processed from mixtures of Clevios PH1000 and DBSA (0.5 v/v %). The highest conductivity, of 600–700 S/cm, is observed for glycerol (5 v/v %), DMSO (5 v/v %), and sorbitol (2.5 wt. %). Considering the lower toxicity of glycerol with respect to DMSO and EG and its ease of process as compared to sorbitol, we employed it as the conductivity enhancing agent in our experiments.

The mixture used to process our films contains 94.5 v/v % of Clevios PH1000, 5 v/v % of glycerol, and 0.5 v/v % of DBSA. Most of the glycerol is expected to be removed after baking, based on water-glycerol temperature-composition phase diagrams obtained at atmospheric pressure.[37] This hypothesis is supported by XPS analysis (shown in Fig. S2), which shows that our films processed from pristine Clevios PH1000 and from Clevios PH1000/glycerol mixtures show similar carbon to sulphur (C/S) and oxygen to sulphur (O/S) ratios. As the addition of glycerol to Clevios PH1000 leads to a significant increase of the amount of C and O in the liquid mixture, a significant presence of glycerol in the film would result in an increase of C/S and O/S ratios, with respect to films processed from pristine Clevios PH1000 (from ~2.7 to ~16 for the C/S ratio and from ~7.5 to ~21 for the O/S ratio). The significant removal of glycerol after baking is further validated by weighing experiments carried out with an analytical balance (see Fig. S3 in supplementary material).[30]

As all organic bioelectronic devices work in aqueous environment, we first investigated the changes of PEDOT:PSS film properties after immersion into deionized water. We found that water immersion induces a considerable decrease in film thickness and, as a consequence, an increase in sheet resistance (see Table S1 in supplementary material[30]). Inspired by this observation, we measured the change in thickness and conductivity of PEDOT:PSS films after immersion for 10 minutes in solvents with different polarity parameters, namely, iso-propanol (polarity parameter 3.9), acetone (5.1), and Milli-Q deionized water (10.2).[38] The polarity parameter, as defined by Snyder,[39] is a measure of the ability of a solvent to interact with various polar test solutes. Interestingly, we observed (Fig. 2) that solvents with higher polarity parameter induce a larger thickness decrease. The decrease ratio is nearly independent of the initial film thickness. The decrease in thickness does not significantly affect the film electrical conductivity (see Table S1 in the supplementary material[30]).

To gain insight into the mechanism of the thickness decrease of PEDOT:PSS films after immersion in water, we analyzed the film chemical composition before and after immersion in water by XPS, focusing in particular on the relative intensities of the characteristic $PEDOT^+$ and PSS^- peaks. Fig. 3(a) shows a portion of the XPS spectra (between 164 and 172 eV binding energy) of PEDOT:PSS films before and after immersion in water. The S(2p) peak at the binding energy of ca. 169 eV corresponds to the sulphur signal of PSS^-, and the doublet peaks at ca.165 eV correspond to the sulphur signal of $PEDOT^+$.[34] Interestingly, after immersion in water, the ratio between the intensities of the PSS^- and $PEDOT^+$ peaks decreases from ca. 2:1 to ca. 1.6:1, which suggests

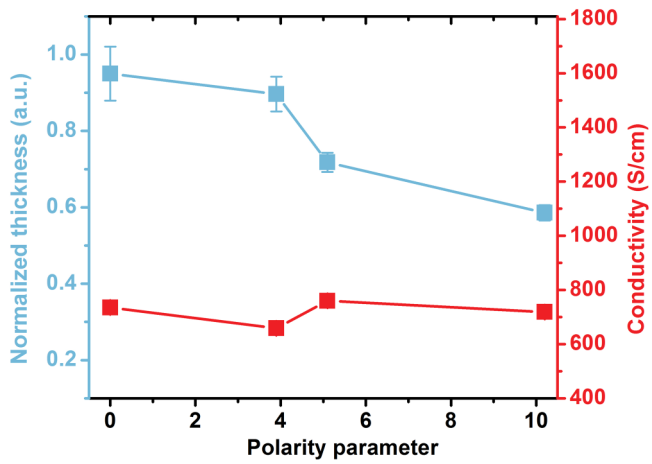

FIG. 2. Thickness and conductivity of PEDOT:PSS films (after immersion for 10 minutes) versus polarity parameters of the solvents: isopropanol (polarity parameter 3.9), acetone (5.1), and Milli-Q deionized water (10.2). The value at polarity parameter =0 corresponds to the film thickness before immersion in the solvents. The thickness values on the left y axes are normalized with respect to the initial films thicknesses (about 150 nm). The PEDOT:PSS film is deposited from a mixture containing 94.5 v/v % of Clevios PH1000, 5 v/v % glycerol, and 0.5 v/v % DBSA. The polarity parameters are obtained from the literature.[38] The error bars correspond to the standard deviations of three samples.

that excess PSS^- is removed from the film, thus explaining the decrease in film thickness.[40] The excess PSS^- is expected to be mostly accumulated on the surface of the films, thus its removal has negligible influence on the film conductivity.[41]

It has been reported that the stability of PEDOT:PSS films in aqueous solutions can be improved by the addition of the cross-linker GOPS, which is expected to prevent partial dissolution or delamination of the PEDOT:PSS films when immersed into an electrolyte solution.[15,42] The detailed mechanism of the cross-linking reaction is still unclear, since several chemical reactions can take place between GOPS and the components of the mixture as well as the glass (SiO_2) substrate. According to the literature, GOPS should react with glycerol via an epoxy-hydroxy (etherification) reaction, to form a network structure.[43,44] Alternative or parallel reaction paths can be the condensation of the methoxy groups with the –OH groups of glycerol[45,46] and at the glass substrate

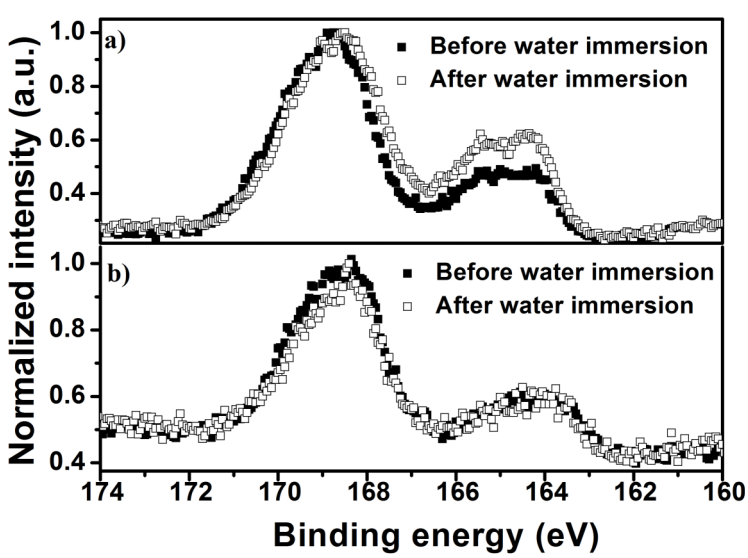

FIG. 3. XPS S(2p) spectra of films processed from mixtures containing 94.5 v/v% of CleviosTM PH1000, 5 v/v% glycerol and 0.5 v/v% DBSA after water immersion for 10 min: (a) without GOPS; (b) with 1 v/v % GOPS.

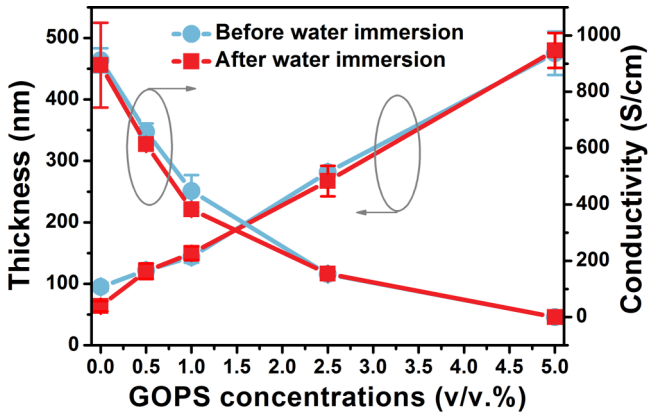

FIG. 4. Thickness (left y scale) and conductivity (right y scale) change of films processed from mixtures containing 94.5 v/v% of CleviosTM PH1000, 5 v/v% glycerol and 0.5 v/v% DBSA with increasing GOPS concentration. The error bar stands for standard deviation of three samples.

surface.[47] It is worth mentioning that we observed a thickness increase and an identical thickness before and after water immersion also for films processed from PEDOT:PSS/GOPS mixtures in the absence of glycerol. This might indicate that the GOPS is also able to react with itself or with PEDOT:PSS, as recently found for the system PEDOT/graphene oxide.[48]

To further clarify the role of GOPS in PEDOT:PSS film, we carried out thickness and resistivity measurements on PEDOT:PSS films with different GOPS contents. Increasing the content of GOPS in the PEDOT:PSS/DBSA/glycerol mixture causes the increase of film thickness and the decrease of film conductivity (Fig. 4). The film thickness increases linearly with the GOPS concentration and reaches 500 nm (i.e., ~5 times the thickness of pristine PEDOT:PSS films) when 5 v/v % GOPS is present in the mixture. This is likely due to the fact that, after the cross linking reaction takes place, a large amount of non-evaporating species (with respect to the PEDOT:PSS initial content of ~1 w/w % in the Clevios PH1000 suspension) is present in the film. To estimate the PEDOT:PSS to cross-linking network ratio, we used the elemental ratios obtained from XPS (see Fig. S2 in the supplementary material[30]). The C/O/Si elemental ratio in the GOPS molecule ($C_9H_{20}O_5Si$) is 9/5/1. Assuming that this ratio is maintained in the film and using the Si content as a reference, we determined the amount of GOPS-related species (cross-linking network) present in the film with respect to PEDOT:PSS. For films processed from a mixture containing 1 v/v % GOPS, the amount of GOPS-related species (C, O, and Si) is ca. 52.5%, which increases to 82.5% for films processed from a mixture containing 2.5 v/v % GOPS. Such a large content of non-evaporating and non-conducting species in the films may indicate that GOPS is able to cross-link with glycerol or other species in the films and explain the increase of thickness and the decrease of electrical conductivity.

Films containing GOPS show no thickness decrease after immersion in water and the corresponding XPS spectra did not show changes neither in PEDOT:PSS ratio (Fig. 3(b)) nor in C/Si ratio (Fig. S2 in the supplementary material[30]). This result proves the effectiveness of the cross-linked network in preventing film dissolution and delamination in aqueous solutions.

As the electrical properties of PEDOT:PSS films are essential for application in OECTs, we tested the electrical stability of the PEDOT:PSS films with and without GOPS in water using a two terminal device. Before the measurements, all films were rinsed in water for 10 min to stabilize the thickness. Although the films containing GOPS show superior mechanical stability, a considerable current decrease is observed during the first 3 h of operation (see Fig. S5 in supplementary material[30]). This phenomenon is likely related to the water absorption by the PSS^- units in the film. The addition of GOPS yields thicker films at a given spin-coating speed. The thicker films likely contain a larger amount of PSS^- units, which are not removed by water due to the presence of the GOPS cross-linker. As a consequence, the excess of hydrophilic PSS^- would absorb larger amounts of water and cause irreversible morphological changes in the film,[49] thus resulting in a decrease in

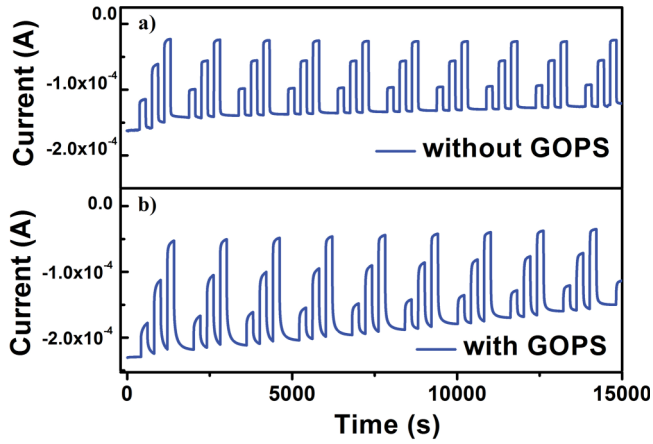

FIG. 5. Transient electrical measurement (10 cycles) with a duration of about 15 000 s (250 min) for OECTs fabricated from PEDOT:PSS/5 v/v % glycerol/0.5 v/v % DBSA films: (a) without GOPS; (b) with 1 v/v % GOPS. The spin-coating speeds for both films are 500 rpm for 10 s followed by 1500 rpm for 45 s. The thickness of the film with and without 1 v/v % GOPS is about 350 nm and 150 nm, respectively.

conductivity. Water absorption of PSS^- likely reaches a saturation state (i.e., PSS^- is fully hydrated) within 3 h and, afterwards, the current becomes stable.

Finally, we studied the effect of GOPS on the electrochemical stability of OECTs by comparison with a device employing no GOPS (Fig. 5). A high surface area (1000–2000 m^2g^{-1}) activated carbon was used as the gate electrode.[50] The source-drain voltage (V_{ds}) was fixed at −0.2 V, and gate bias (V_g) was pulsed from 0 to 0.6 V with a step of 0.2 V. Each OECT was run for 10 cycles for a total time of about 250 min. As shown in Fig. 5, for films processed at the same spin-coating speeds, PEDOT:PSS channel containing GOPS shows a higher current (I_{ds}), which should result from the higher film thickness. OECTs with and without GOPS crosslinker show comparable electrochemical stability after 10-cycles operation. However, devices containing GOPS show a considerable negative base current shift (Fig. 5(b)), which is likely caused by the water absorption of PSS^- in the films, and by the presence of the non-conductive species in the cross-linked network.

In summary, we have investigated the variations of the properties of PEDOT:PSS films in liquids with different polarities. Increasing the solvent polarity leads to a decrease in thickness and an increase in sheet resistance, while the film electrical conductivity remains substantially unaffected. XPS analysis reveals that the thickness decrease is caused by the removal of PSS^- from the film. The thickness decrease after solvent immersion can be minimized when a GOPS crosslinker is added to the processing mixture. OECT based on PEDOT:PSS film processed with or without GOPS crosslinker shows comparable current/voltage characteristics.

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[1] G. Malliaras and R. Friend, Phys. Today **58**(5), 53–58 (2005).
[2] F. Cicoira and C. Santato, *Organic Electronics: Emerging Concepts and Technologies* (John Wiley & Sons, 2013).
[3] C. W. Tang, Appl. Phys. Lett. **48**(2), 183–185 (1986).
[4] G. Horowitz, Adv. Mater. **10**(5), 365–377 (1998).
[5] M. Berggren and A. Richter-Dahlfors, Adv. Mater. **19**(20), 3201–3213 (2007).
[6] H. S. White, G. P. Kittlesen, and M. S. Wrighton, J. Am. Chem. Soc. **106**(18), 5375–5377 (1984).
[7] D. Nilsson, T. Kugler, P.-O. Svensson, and M. Berggren, Sens. Actuators, B **86**(2), 193–197 (2002).
[8] D. A. Bernards, D. J. Macaya, M. Nikolou, J. A. DeFranco, S. Takamatsu, and G. G. Malliaras, J. Mater. Chem. **18**(1), 116–120 (2007).
[9] J. Isaksson, P. Kjäll, D. Nilsson, N. Robinson, M. Berggren, and A. Richter-Dahlfors, Nat. Mater. **6**(9), 673–679 (2007).

[10] H. Tang, F. Yan, P. Lin, J. Xu, and H. L. Chan, Adv. Funct.Mater. **21**(12), 2264–2272 (2011).
[11] G. Tarabella, C. Santato, S. Y. Yang, S. Iannotta, G. G. Malliaras, and F. Cicoira, Appl. Phys. Lett. **97**(12), 123304 (2010).
[12] D. A. Bernards and G. G. Malliaras, Adv. Funct. Mater. **17**(17), 3538–3544 (2007).
[13] P. Lin, F. Yan, and H. L. Chan, ACS Appl. Mater. Interfaces **2**(6), 1637–1641 (2010).
[14] F. Cicoira, M. Sessolo, O. Yaghmazadeh, J. A. DeFranco, S. Y. Yang, and G. G. Malliaras, Adv. Mater. **22**(9), 1012–1016 (2010).
[15] S. Yoonáyang, Chem. Commun. **46**(42), 7972–7974 (2010).
[16] H. Tang, P. Lin, H. L. Chan, and F. Yan, Biosens. Bioelectron. **26**(11), 4559–4563 (2011).
[17] P. Lin, X. Luo, I. Hsing, and F. Yan, Adv. Mater. **23**(35), 4035–4040 (2011).
[18] M. H. Bolin, K. Svennersten, D. Nilsson, A. Sawatdee, E. W. Jager, A. Richter-Dahlfors, and M. Berggren, Adv. Mater. **21**(43), 4379–4382 (2009).
[19] L. H. Jimison, S. A. Tria, D. Khodagholy, M. Gurfinkel, E. Lanzarini, A. Hama, G. G. Malliaras, and R. M. Owens, Adv. Mater. **24**(44), 5919–5923 (2012).
[20] D. Khodagholy, T. Doublet, P. Quilichini, M. Gurfinkel, P. Leleux, A. Ghestem, E. Ismailova, T. Hervé, S. Sanaur, and C. Bernard, Nat. Commun. **4**, 1575 (2013).
[21] S. Y. Yang, J. A. DeFranco, Y. A. Sylvester, T. J. Gobert, D. J. Macaya, R. M. Owens, and G. G. Malliaras, Lab Chip **9**(5), 704–708 (2009).
[22] K. Tybrandt, R. Forchheimer, and M. Berggren, Nat. Commun. **3**, 871 (2012).
[23] D. Khodagholy, J. Rivnay, M. Sessolo, M. Gurfinkel, P. Leleux, L. H. Jimison, E. Stavrinidou, T. Herve, S. Sanaur, R. M. Owens, and G. G. Malliaras, Nat. Commun. **4**, 2133 (2013).
[24] J. Ouyang, Q. Xu, C.-W. Chu, Y. Yang, G. Li, and J. Shinar, Polymer **45**(25), 8443–8450 (2004).
[25] B. D. Martin, N. Nikolov, S. K. Pollack, A. Saprigin, R. Shashidhar, F. Zhang, and P. A. Heiney, Synth. Met. **142**(1), 187–193 (2004).
[26] A. Elschner and W. Lövenich, MRS Bull. **36**(10), 794–798 (2011).
[27] E. Stavrinidou, P. Leleux, H. Rajaona, D. Khodagholy, J. Rivnay, M. Lindau, S. Sanaur, and G. G. Malliaras, Adv. Mater. **25**(32), 4488–4493 (2013).
[28] L. Kergoat, B. Piro, D. T. Simon, M. C. Pham, V. Noël, and M. Berggren, Adv. Mater. **26**(32), 5658–5664 (2014).
[29] F. Smits, Bell Syst. Tech. J. **37**(3), 711–718 (1958).
[30] See supplementary material at http://dx.doi.org/10.1063/1.4905154 for the film conductivitites with sorbitol as conductivity enhancing agent, XPS, analytical balance weighing, Fourier transform spectroscopy, and two-terminal electrical measurements.
[31] J. Kim, J. Jung, D. Lee, and J. Joo, Synth. Met. **126**(2), 311–316 (2002).
[32] F. Zhang, T. Nyberg, and O. Inganäs, Nano Lett. **2**(12), 1373–1377 (2002).
[33] J. Ouyang, C. W. Chu, F. C. Chen, Q. Xu, and Y. Yang, Adv. Funct.Mater. **15**(2), 203–208 (2005).
[34] Y. H. Kim, C. Sachse, M. L. Machala, C. May, L. Müller-Meskamp, and K. Leo, Adv. Funct. Mater. **21**(6), 1076–1081 (2011).
[35] S. Jönsson, J. Birgerson, X. Crispin, G. Greczynski, W. Osikowicz, A. D. Van Der Gon, W. R. Salaneck, and M. Fahlman, Synth. Met. **139**(1), 1–10 (2003).
[36] N. Kim, S. Kee, S. H. Lee, B. H. Lee, Y. H. Kahng, Y. R. Jo, B. J. Kim, and K. Lee, Adv. Mater. **26**(14), 2268–2272 (2014).
[37] M. Oliveira, A. Teles, A. Queimada, and J. Coutinho, Fluid Phase Equilib. **280**(1), 22–29 (2009).
[38] P. C. Sadek, *The HPLC Solvent Guide* (John Wiley & Sons, 2002).
[39] L. Snyder, J. Chromatogr. A **92**(2), 223–230 (1974).
[40] D. A. Mengistie, M. A. Ibrahem, P.-C. Wang, and C.-W. Chu, ACS Appl. Mater. Interfaces **6**(4), 2292–2299 (2014).
[41] G. Greczynski, T. Kugler, and W. Salaneck, Thin Solid Films **354**(1), 129–135 (1999).
[42] L. Kergoat, B. Piro, D. T. Simon, M.-C. Pham, V. Noël, and M. Berggren, Adv. Mater. **26**(32), 5658–5664 (2014).
[43] S. Doszlop, V. Vargha, and F. Horkay, Chem. Eng. **22**(3), 253–275 (1978).
[44] J.-P. Pascault and R. J. Williams, *Epoxy Polymers* (John Wiley & Sons, 2009).
[45] W. Noll, *Chemistry and Technology of Silicones* (Elsevier, 1968).
[46] M. A. Brook, Y. Chen, K. Guo, Z. Zhang, W. Jin, A. Deisingh, J. Cruz-Aguado, and J. D. Brennan, J. Sol-Gel. Sci. Technol. **31**(1-3), 343–348 (2004).
[47] A. K. Wong and U. J. Krull, Anal. Bioanal. Chem. **383**(2), 187–200 (2005).
[48] V. C. Tung, J. Kim, L. J. Cote, and J. Huang, J. Am. Chem. Soc. **133**(24), 9262–9265 (2011).
[49] A. M. Nardes, M. Kemerink, M. De Kok, E. Vinken, K. Maturová, and R. Janssen, Org. Electron. **9**(5), 727–734 (2008).
[50] J. Sayago, F. Soavi, Y. Sivalingam, F. Cicoira, and C. Santato, J. Mater. Chem. C **2**, 5690 (2014).
[51] $\rho/d = 4.5324 \times V/I$. Where ρ is the resistivity of the film, d is the film thickness, I is the supplied current, and V is the measured voltage.[29] The conductivity σ then can be extracted using the following relation: $\sigma = 1/\rho$