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Highly stretchable PEDOT:PSS organic electrochemical transistors achieved via polyethylene glycol addition

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Abstract

Organic electrochemical transistors (OECTs) are widely used in biosensing and bioelectronics, due to their ability to convert ionic signals into electronic signals and their high transconductance. Stretchable OECTs are particularly suited for on-skin and on-organ bioelectronics, since they are able to record or transmit signals under mechanical strain. Most stretchable OECTs are based on the conducting polymer poly(3, 4-ethylenedioxythiophene) doped with polystyrene sulfonate (PEDOT: PSS), which needs to be appropriately processed to yield stretchable films. Here we report stretchable OECTs that are obtained by modifying the mechanical properties of PEDOT:PSS films via the addition of low-molecular weight polyethylene glycol (PEG), which acts as a plasticizer. The presence of PEG in the films prevents the formation of cracks under strain while maintaining a high electrical conductivity, thus resulting in improved electromechanical properties. In particular, the additon of PEG leads to a higher channel thickness and increased ion mobility in the films, thus resulting in stretchable OECTs with high transconductance and fast response time. This work shows that high stretchability, high transconductance and fast repsonse time can be simultaneously obtained in OECTs, paving the way for their applications in conformable devices at the human-machine interface.

1. Introduction

Increasing demand for intelligent, wearable and integrated electronics has driven the unprecedented interest in stretchable electronic devices, such as sensors [1], transistors [2], solar cells [3], batteries and capacitors [4, 5]. Organic electrochemical transistors (OECTs) are already widely used in bioelectronics, where a combined electronic-ionic transport is required [6–9]. Stretchable OECTs are highly demanded for on-skin or on-organ applications because of their ability to show strain-insensitive electric performance.

Currently, most OECTs use thin films of the conducting polymer poly(3, 4-ethylenedioxythiophene) doped with polystyrene sulfonate (PEDOT:PSS) as channel material, because of its high conductivity, biocompatibility, ease of process and stability in water at a physiological pH [10, 11]. As PEDOT:PSS films are, in general, inherently brittle, several approaches have been proposed to improve their mechanical properties. in order to incorporate them in stretchable devices. Stretchable PEDOT:PSS-based OECTs have been demonstrated by our group via a combination of parylene transfer, orthogonal patterning and buckling of PEDOT:PSS films on a pre-stretched elastomer. These devices showed almost unchanged transistor performance up to 30% strain [12]. An alternative approach used a laser patterning method to fabricate stretchable metallic interconnections on elastomers to yield OECTs with a maximum stretchability of 38% while maintaining a transconductance as high as 0.35 mS [13]. Matsuhisa *et al* fabricated stretchable Au conductors controlling the strain-induced microcrack propagation in Au thin films by varying the evaporation rate. These Au conductors were utilized to fabricate stretchable OECTs, based on PEDOT:PSS, with a high transconductance both at 0% strain (0.54 mS) and 140% strain (0.14 mS) [14]. Recently, our group also demonstrated stretchable OECTs on PDMS, able to maintain stable performance up to 30% strain, by decreasing the thickness of PEDOT:PSS films to ~50 nm and using a baking temperature to 100 °C [15].

Decreasing film thickness is a well-known method to obtain better stretchability of thin films of conducting polymers or metals on elastomers, due to a transition from a 3D packing to a 2D percolating structureand a reduced mechanical mismatch with the substrate [14–18]. However, as the transconductance of OECTs is propotional to the channel thickness [6], a compromise must be reached to balance the electrical and mechanical properties of the films.

Another widely-used approach to yield stretchable films is to modify the mechanical properties conducting polymers by blending them with plasticizers, such as Capstone FS-30 (Zonyl) [19, 20], Triton X-100 [21] and ionic liquids [22], whose effect is to decrease the Young's modulus and to increase the elongation at the break. Low-molecular-weight polyethylene glycol (e.g. PEG 400) has been proven to act as a plasticizer in polymer processing, e.g. for poly(lactic acid), by decreasing the Young's modulus and increasing the elongation at the break [23–26]. In addition, when blended with electrolytes, PEG has shown to lead to an improvement of ion transport [27]. Moreover, low-molecular-weight PEG shows high aqueous solubility, antifouling properties [28], and a moderate ability to enhance conductivity of PEDOT:PSS [29]. This ensemble of properties makes it an interesting candidate as plasticizer for applications in bioelectronic devices based on PEDOT:PSS.

In this work, we achieved highly stretchable OECTs by using PEG 400 as plasticizer for PEDOT:PSS films. The PEG 400 remains inside the films after baking, and improves the electromechanical properties by limiting the formation of cracks under strain. This allowed us to fabricate high transconductance OECTs keeping similar performance between 0% and 45% strain. As PEG also favours ion transport in the films, our OECTs showed a faster response time compared to reference devices.

2. Experimental

2.1. Materials

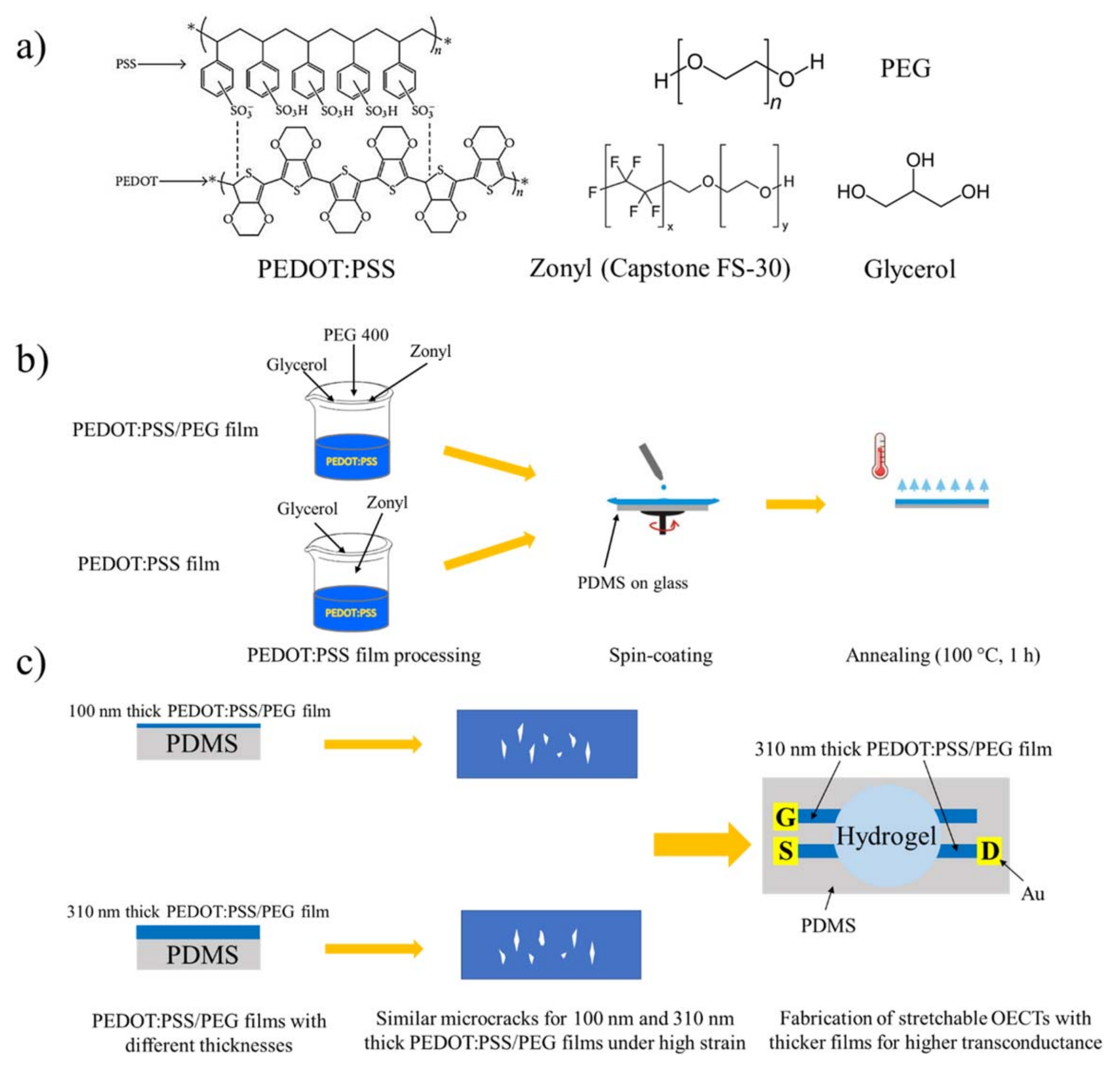
The PEDOT:PSS aqueous suspension (Clevios PH1000) was purchased from Heraeus Electronic Materials GmbH (Leverkusen, Germany). PEG 400, the liquid metal Gallium-Indium eutectic (EGaIn) 495425 and Capstone FS-30 (Zonyl) were purchased from Sigma-Aldrich. Glycerol (99.5 + % purity) was purchased from Caledon Laboratories Ltd (Georgetown, ON). The Orthogonal photoresist kit was supplied by Orthogonal Inc. Glass slides were purchased from Corning. Polydimethylsiloxane (PDMS, Sylgard 184 silicone elastomer kit) was purchased from Dow Corning.

2.2. PEDOT:PSS film preparation

The PEDOT:PSS/PEG films were deposited onto PDMS substrates by spin coating a mixture of Clevios PH1000, 4 v/v% PEG 400, 1 v/v% fluorosurfactant Capstone FS-30 and 5 v/v% glycerol. Control samples without PEG 400 were also prepared. The two kinds of samples described above are named as PEDOT:PSS/PEG and PEDOT:PSS films. The PDMS substrates (160 μm thick) were cleaned by exposure to UV/O_3 for 20 min. Among all PEGs with different molecular weights, PEG 400 was selected as plasticizer for PEDOT:PSS due to its liquid form at room temperature and ease of process. After deposition, the films were baked at 100 °C for 1 h. The film thickness was measured with a profilometer (Dektak 150) using a 12.5 μm stylus tip with a 10 mg force, after transferring them from PDMS onto glass using a water-soluble tape (3 M 5414). We observed thicknesses of ~300 nm (1000 rpm), and ~100 nm (4000 rpm) for PEDOT:PSS/PEG films and ~50 nm (4000 rpm) for PEDOT:PSS films. The difference in thickness for films deposited at the same spin coatng speed is due to the presence of PEG in the films after baking.

2.3. Characterization of films

The optical microscopy images were obtained with a Carl Zeiss AX10 microscope. The electromechanical properties of the films were evaluated by measuring the change of the current flowing in the films as a function of time during stretching and release cycles at different applied tensile strain percentage [30]. The latter is defined as $[(L' - L)/L] \times 100\%$, where L and L′ denote the relaxed and stretched lengths of the PDMS substrate, respectively. Two EGaIn *contacts* were applied at the extremities of the films to facilitate the electrical contact. The current was normalized with respect to the value measured for unstretched films. As figure of merit to assess the electromechanical properties of the films, we used the *ratio* $\Delta I/I_{0x}$, where ΔI indicates the current variation between the stretched and released state, and I_{0x} is the current in the released state after the first application of x% strain (pre-set strain). A low value of $\Delta I/I_{0x}$ indicates that the electrical properties are not significantly affected by mechanical deformation, which is a desirable property for stretchable OECTs [15]. Scanning electron microscopy (SEM) measurements were performed with a JEOL JSM-7600TFE Field Emission Scanning Electron Microscope (FE-SEM). X-ray photoelectron spectroscopy (XPS) was performed using a VG ESCALAB 3MKII system with Mg-Ka x-ray source in an ultra-high vacuum.



Scheme 1. a) Molecular structures of PEDOT:PSS, glycerol, fluorosurfactant (Zonyl) and PEG, b) schematic overview of the sample preparation steps for PEDOT:PSS films and PEDOT:PSS/PEG films and c) schematic overview of the fabrication of stretchable OECTs with thick PEDOT:PSS/PEG films

2.4. Polyampholyte hydrogel electrolyte synthesis

The polyampholyte hydrogel synthesis has been described previously [31]. Briefly, 1 M Sodium 4-vinylbenzenesulfonate (NaSS) and 1 M [3-(methacryloylamino)propyl] trimethylammonium chloride (MPTC) with 4,4-Azobis(4-cyanovaleric acid) (thermal initiator, 0.25 mol%, respect to the amount of NaSS and MPTC) were dissolved in deionized water to form the precursor solution. The precursor solution was heated at 70 °C in the convection oven for 6 h to yield the as-prepared hydrogel. Then, the as-prepared hydrogel was purified in deionized water to remove unreacted monomers. The purified hydrogel was further dialyzed in the electrolyte solution (0.1 M NaCl solution) to let ions diffuse inside the hydrogel.

2.5. OECT fabrication and characterization

OECTs were fabricated following our previously reported procedure: Au contact pads (thickness ~20 nm with a ~2 nm Ti adhesion layer) were patterned via parylene transfer and the PEDOT:PSS channels with orthogonal photolithography [12, 15]. As the electrolyte, we used either a 0.1 M NaCl aqueous solution confined in an elastic well made of stretchable tape (3 M VHB 4905) or a cut and paste polyampholyte hydrogel containing NaCl. As the gate electrode we used activated carbon on carbon paper for devices using aqueous electrolytes [30] and a planar PEDOT:PSS stripe for those using a hydrogel electrolyte. Liquid metal EGaIn was applied at the source-drain electrodes to facilitate the probing. The transistor electrical characteristics were measured with an Agilent B2900A source measure unit controlled with Quick IV Measurement software, and the strain was applied *in situ* with a LabVIEW software controlled tensile tester.

3. Results and discussions

The molecular structures of the chemicals used in this work as well as the preparation steps of films and devices are shown in scheme 1. The films obtained

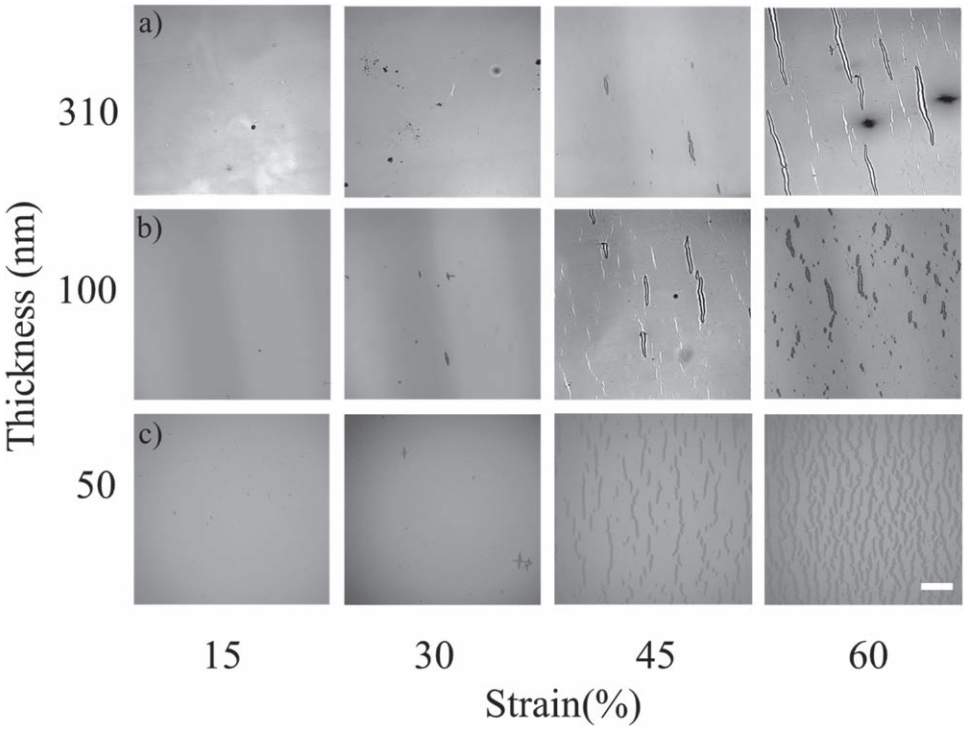


Figure 1. Optical microscopy images under strain of ~300 nm thick PEDOT:PSS/PEG film (a), ~100 nm thick PEDOT:PSS/PEG film (b) and ~50 nm thick PEDOT:PSS film on PDMS substrate. All films were baked at 100 °C for 1 h. The scale bar is 200 μm.

from mixtures containing Clevios PH 1000 and 4 v/v% PEG 400 show a conductivity of ~200 S cm^{-1}, which increases up to ~450 S cm^{-1} upon addition of 5 v/v% glycerol and 1 v/v% Capstone.

The optical microscopy images of PEDOT:PSS (~50 nm thick) and PEDOT:PSS/PEG (~100 and ~300 nm thick) films under 15%, 30%, 45% and 60% strains are shown in figure 1. No significant crack formation is observed at 15% and 30% strain for all films. When the strain reaches 45%, a high density of cracks is observed on ~50 nm thick PEDOT:PSS films (figure 1(c)). As shown by prelimimary SEM investigation, the cracks propagate through the entire film thickness (figure S1 is available online at stacks.iop.org/FPE/4/044004/mmedia). The length and density of cracks increase at the application of 60% strain, in accordance with our previous study [15]. The addition of PEG significantly affects the formation of cracks: both ~100 and ~300 nm thick PEDOT:PSS/PEG films show short and sparse cracks up to 45% and 60% strain. Interestingly, increasing the film thickness does not impact the density and the length of the cracks, unlike to what we previously observed for PEDOT:PSS films without PEG [15].

The shape and density of cracks have an important effect on the current flowing in the films under strain (figure 2(a)) and on the $\Delta I/I_{0x}$ (figure 2(b)). In absence of cracks, or at low crack density, there is a small difference in current between the stretched and released state, which results in a small $\Delta I/I_{0x}$. Long and high density cracks partially interrupt the electrical conducting path during stretching, resulting in a drop of the current. When the strain is released, the cracks re-connect in part and the current is partially recovered, thus resulting in a large $\Delta I/I_{0x}$. The ~50 nm thick PEDOT:PSS films maintain a small $\Delta I/I_{0x}$ (less than 0.05) under 15% and 30% strain. At strain of 45% or larger, there is a clear increase of $\Delta I/I_{0x}$ (figure 2(b)), which reaches almost 1 at 75% applied strain. The addition of PEG clearly improves the electromechanical properties of the films, by leading to a smaller $\Delta I/I_{0x}$, which remains below 0.1 up to 60% strain. At strains of 75% or higher the $\Delta I/I_{0x}$ increases up to a value of about 0.8 at 120% strain.

We have recently shown that the electromechanical properties of PEDOT:PSS films strongly depend on the thickness [15]. Thin films (~50 nm) show better electromechanical characteristics than thick ones (~130 nm), due a lower density of strain-induced cracks [15]. However, for OECT fabrication, a high film thickness would be preferred to maximize the device transconductance [6]. The effect of PEDOT:PSS/PEG film thickness on the electromechanical properties is shown in figure 2(c). At 15% strain, the normalized current of ~300 nm thick film remains unchanged while that of ~100 nm film drops by 15%. At 30% and 45% strain the ~300 nm film still maintain 90%, and 75% of the initial current. Notably, the $\Delta I/I_{0x}$ value of ~300 nm film is similar but slightly lower than that of ~100 nm film when the applied strain is 45% or less (figure 2(d)). When the strain reaches 60%, the current of both ~300 and ~100 nm

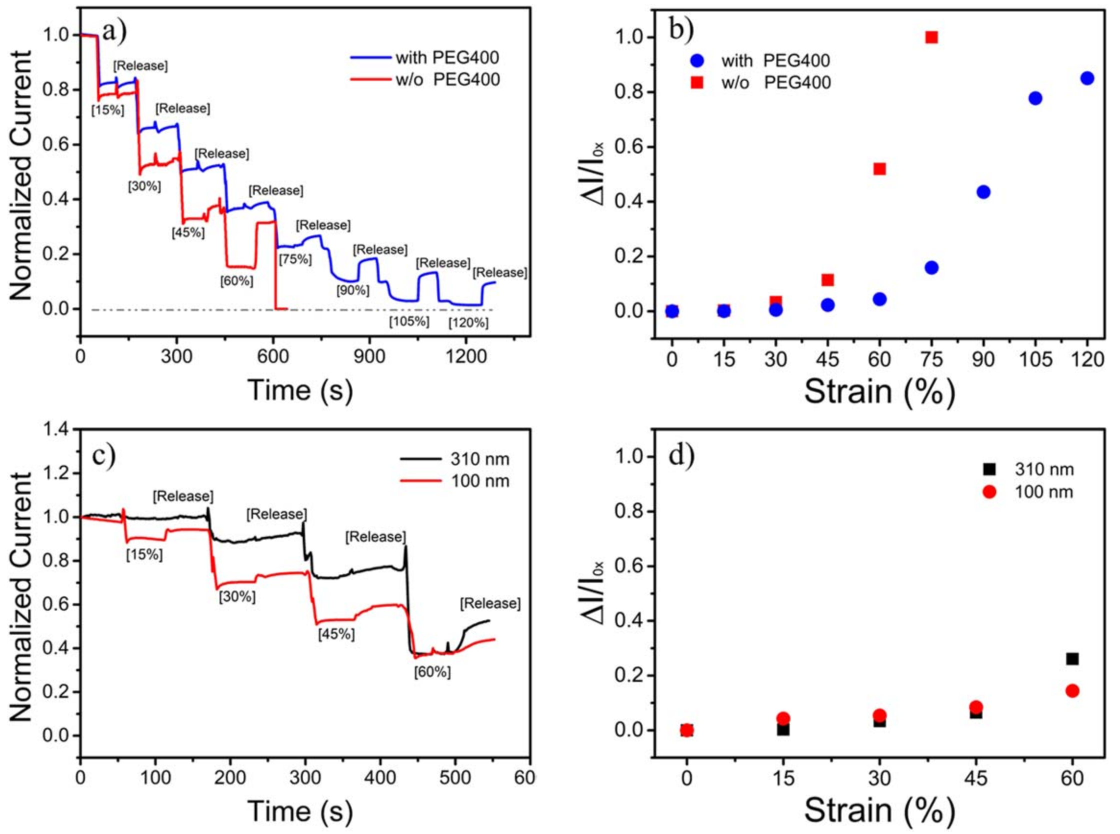


Figure 2. Normalized current versus time at different applied strain percentages (a) and $\Delta I/I_{0x}$ versus strain percentage (b) for PEDOT:PSS/PEG and PEDOT:PSS films on PDMS. The thickness is ~50 nm for PEDOT:PSS film and ~100 nm for PEDOT:PSS/PEG films. Same plots for PEDOT:PSS/PEG films with ~100 and ~300 nm thickness (c) and (d). A constant voltage of 0.2 V is applied. The applied strain is increased in 15% per step and the strain rate is 2.5%/s. For each cycle, the samples are kept in the stretched and released state for 50 s. All films were baked at 100 °C for 1 h. In each experiment, at least 4 films were measured to ensure the results are repeatable.

films drops to 40% of the initial value, and ~100 nm film exhibits a smaller $\Delta I/I_{0x}$ value. Since increasing the thickness of PEDOT:PSS/PEG films from ~100 to ~300 nm does not significantly affect the $\Delta I/I_{0x}$ value, we fabricated OECTs using ~300 nm PEDOT:PSS/PEG films to achieve larger transconductance without sacrificing the electromechanical properties.

The enhanced stretchability of PEDOT:PSS/PEG films is attributed to the plasticizing effect of PEG, which remains in the film after baking. We investigated the composition of PEDOT:PSS and PEDOT:PSS\PEG films by XPS. Clear shifts in C(1s) and O(1s) core level spectra indicates the presence of PEG 400 (figure S2) [29]. This is consistent with the higher thickness of PEDOT:PSS/PEG films with respect to PEDOT:PSS ones processed in the same conditions. According to a previous study [32], the PEG remaining in the films functions as a soft matrix, with PEDOT:PSS particles embedded on it. The hydrogen bonds between PEG and PSS modifies the interaction between PEDOT and PSS, leading to the enhanced stretchability [32]. Overall, when PEG is present in the film, the adverse effect of increasing film thickness on electromechanical properties is compensated by improved plasticity.

To gain more insight on the stabilty of ~300 nm PEDOT:PSS/PEG film, a stretch-release cycle test at 30% and 45% strain was performed. The samples were stretched for 50 s and released for the same amount of time for 100 times. The film resistance, calculated through the applied voltage and measured current, increases during the first of three stretch-release cycles, then it reaches a stable value, that is maintained up to 100 cycles for both 30% and 45% strain (figure 3(a)). The result demonstrates an excellent cyclic stability towards stretch-release cycles, which can be explained by the fact that the shape and density of the cracks do not change significantly between the 3rd and the 100th stretch-release cycles (figure 3(b)).

Stretchable OECTs were fabricated on PDMS according to a method previously reported by our group [12, 15]. We used a stretchable polyanpholyte hydrogel or an aqueous electrolyte as the gating medium. To facilitate probing on the tensile tester, large devices were used (channel length, L, of 8 mm and a width, W, of 2 mm). On the bases of the results shown in figure 3, we pre-stretched the PEDOT:PSS/PEG films by applying 45% strain for three times to reach a stable current.

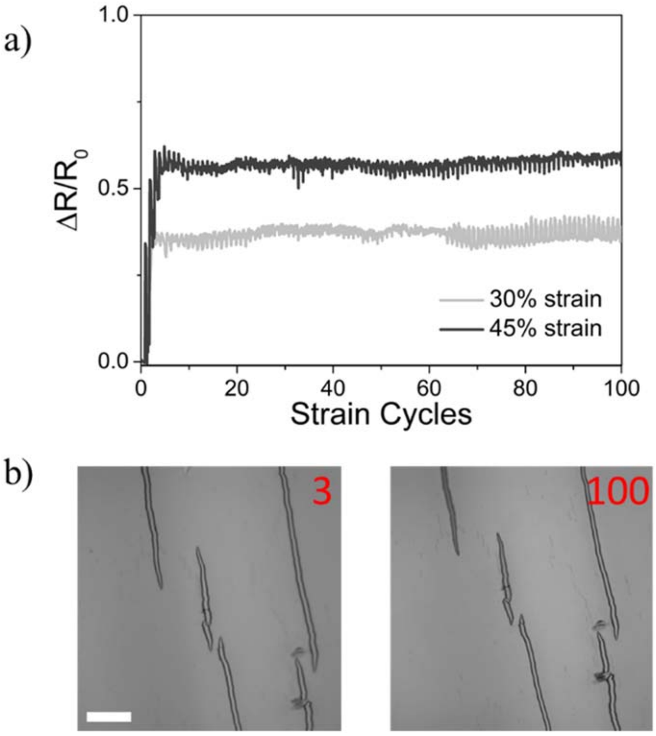


Figure 3. Resistance change during 100 stretch-release cycles from 0% to 30% and 0% to 45% strain for PEDOT:PSS/PEG film (~300 nm thick) on PDMS (a). Density and shape od cracks for a ~300 nm thick PEDOT:PSS/PEG film (100 °C baking for 1 h) after 3 and 100 strain cycles at 45% strain (b). R_0 is the initial resistance of PEDOT:PSS/PEG film before stretching. The scale bar is 200 μm.

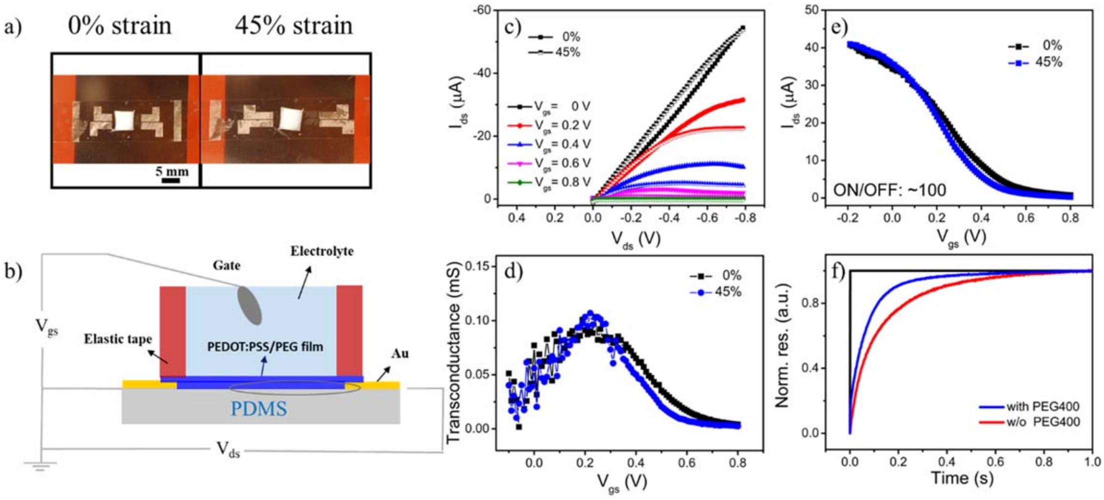


Figure 4. Optical images of stretchable OECTs using PEDOT:PSS/PEG channel and gate electrode and polyampholyte hydrogel containing NaCl electrolyte at 0% and 45% strain (a); schematic image of device structure of stretchable OECTs using an aqueous electrolyte and an activated carbon gate (b). Electrical characteristic of stretchable OECTs based on PEDOT:PSS/PEG films on PDMS, using an activated carbon gate electrode and 0.1 M NaCl aqueous solution as electrolyte (c)–(f): output curves (c), transconductance (d) and transfer curves (e) at 0% and 45% strain, normalized time response of I_{ds} during de-doping ($V_g = 0.4$ V) in presence and in absence of PEG 400 into the films (f). The black step indicates the pulse of 0.4 V gate voltage. All films were baked at 100 °C for 1 h. The thicknesses of PEDOT:PSS/PEG and PEDOT:PSS film are ~300 nm and ~130 nm, respectively. The channel length and width are 8 and 2 mm.

For the hydrogel-gated OECTs (video S1), a planar PEDOT:PSS stripe parallel to the channel was used as the gate (figure 4(a)). These devices show similar transfer characteristics when operated at 0% and 45% strain (figure S3). These results demonstrate that our hydrogel electrolyte can efficiently gate PEDOT:PSS/PEG film at different strains. However, the low mobility of ions inside the hydrogel limits the performance of OECTs (ON/OFF ratio ~5).

To achieve better device performance, we also fabricated stretchable OECTs using an aqeous electrolyte (0.1 M NaCl solution) confined in an elastic well made of stretchable tape (3 M VHB 4905). Activated carbon on carbon paper was used as the gate electrode (figure 4(b)) [30]. The output and transfer characteristics (figures 4(c), (e)) show that the devices performance is similar at 0% and 45% strain. These property makes our devices suitable for on-skin bioelectronic (30% strain) and for application where larger strains are required, such as heart-beat monitoring electronics and sensors applied on joints or knees [33]. Four devices were measured to calculate the transconductance and ON/OFF ratio. The values of ON/OFF ratio range from 50 to 160. We extracted a maximum transconductance of about 0.1 mS ($V_{gs} = 0.2$ V, figure 4(d)) and an average ON/OFF ratio of 100 (I_{ds} ($V_{gs} = 0$ V)/I_{ds} ($V_{gs} = 0.8$ V), figure 4(e)), both higher compared to devices with the same geometry using PEDOT:PSS films without PEG, which show a transconductance of ~0.07 mS and an ON/OFF ratio of ~10 (figure S4) [15]. The low ON/OFF ratio of OECTs on PDMS are likely due to impurities of unreacted momomers present at PDMS surface and to an irregular morphology [34, 35]. The beneficial effect of PEG on the ON/OFF ratio is still under investigation. However, we hypothesize that the PEG present in the film prevents or reduces the diffusion of unreacted momomer towards the conducting PEDOT regions, thus reducing its impact on the electrical properties. Interestngly, OECTs based on PEDOT:PSS/PEG channels also show a faster response time than those based on PEDOT:PSS (figure 4(f)), even at high channel thickness (~300 nm). This is likely because the presence of glycol groups in PEG 400 facilitates the mobility of cations [36, 37].

4. Conclusions

In this work, we showed that the addition of PEG 400 to PEDOT:PSS processing mixtures strongly improves the electromechanical properties of films on PDMS substrates. When used as channels in OECTs, such films result in a higher stretchability and improved electrical performance. Optical microscopy images show that the presence of PEG significatly reduces the formation of cracks in the films, thus resulting in small vatiation of the film resistance up to 45% strain. Interestingly, increasing film thickness from ~100 to ~300 nm does not deteriorate the electromechanical properties. Due to the presence of hydyoxyl groups, which likely facilitate ion transport, these stretchable OECTs also show faster response time compared to devices without PEG. This work provides an easy method to enhance the electro mechanical properties of conducting polymer films and paves the way for high performace stretchable OECTs for applications as conformable biosensors and bio-electronic devices.

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