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Published in:
Small

DOI:
10.1002/smll.201803939

Publicerad: 01/01/2019

Please cite the original version:

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Stretchable High-Mobility and Multi-Functional Transistors with Intrinsic Self-Healing Properties of all Components

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Flexible and stretchable field-effect transistor (FET) is an essential element in various modern electronics.\textsuperscript{1,2} To realize the potential of these devices in harsh and real-world conditions and to extend its application spectrum, there is a need to introduce new functionalities into the device elements. Here, we report on solution-processable elements that empower flexible and stretchable FETs with high hole-mobility ($\mu_h \sim 10$ cm$^2$ V$^{-1}$ s$^{-1}$) and relatively low operating voltages (<8V) as well as with self-healing properties of all device components. The device exhibits repeatable intrinsic and autonomic self-healing ability, viz. without use of any external trigger, enabling the restoration of its electrical and mechanical, both after micro-scale damages or complete cut of the device, by, for example, scissors. The device can be repeatedly stretched for more than 200 cycles of up to 50% strain without a significant loss in electrical properties. The device, in the form of a 3 $\mu$m-thick substrate-free skin tattoo, exhibits multi-functional sensing properties, such as detection of temperature and humidity. With this unprecedented smart transistor, we demonstrate that self-healing polymers can offer more than what we thought by allowing the biomimetic electronics.

Soft and flexible devices offer a wide spectrum of new promising applications.\textsuperscript{3-12} For example; epidermal sensors that can continuously monitor the body, such as temperature, sweat, and motion, and deliver information regarding health status would revolutionize present prognosis methods by introducing fast, cheap, and non-invasive alternative.\textsuperscript{3,8,13,14} Soft and flexible sensors must satisfy several mechanical requirements, including conformal attachment to the body,
stretchability and softness, to be utilized with the highest efficiency. However, devices introducing these properties are susceptible to mechanical/structural damages, such as cracks and scratches, which are induced by the combined effect of their soft nature and their incompatibility in mechanical stress with human skin. As a result, this leads to a lower durability, decreased lifetime, and damaged performance in many cases. To address this issue, one can mimic biological systems by introducing the self-healing capability, which is a vital property for many organisms in nature, into the flexible and soft devices allowing the recovery of damages without any external intervention.

Excellent progress in the development of new self-healing materials has been introduced in non-electronic systems as well as in (chemi)resistors, supercapacitors, and electrochemical devices. FETs, which offers considerable advantages as a sensor over competing strategies, such as electrochemical and optical based sensors, by the capability of delivering a label-free response using a simple electronic read-out set-up that can be easily miniaturized by employing printed circuit technologies, has not been targeted yet. For the preparation FETs, self-healing dielectric, conductive, and semiconductive materials are required, with the latter being the most challenging one. The difficulty of obtaining such material arises from the rigidity of semiconductors because of their conjugated and high crystalline structure, which is contradictory to the softness and high chain mobility of self-healing materials. Recently, Bao and coworkers have succeeded to synthesize a healable semiconducting polymer based on 3,6-di(thiophen-2-yl)-2,5-dihydropyrrolo[3,4-c]pyrrole-1,4-dione (DPP) repeating units and non-conjugated 2,6-pyridine dicarboxamide (PDCA) moieties with a $\mu_n$ of $\sim$1.4 cm$^2$ V$^{-1}$ s$^{-1}$, $\sim$10$^6$ on/off ratio, and high operating voltages, up to -60 V. Nevertheless, the healing process needed solvent treatment or high temperatures for electrical and mechanical recovery and the self-healed damage size was very small (nanocracks). Herein, we introduce the preparation of a self-healing semiconductor as well as its integration into intrinsic and fully self-healing FET that exhibits high electrical performances and recover its structural and electrical after micro-sized damages as well as complete cut of the device. We show that the self-healing merit of the FET is also preserved when the device is used for sensing physical (e.g., temperature) or chemical (e.g., humidity) stimuli.

An intrinsic self-healing disulfide-containing poly(urea-urethane) (PU) which is similar to previously reported polymers was used in this work. It is composed of two monomers,
4-aminophenyl disulfide (APDS) and poly(propylene glycol) tolylene 2,4-diisocyanate terminated (PPG-TDI) and contains dynamic hydrogen and disulfide bonds. The reversible nature of these bonds combined with the low glass transition temperature ($T_g$) of the polymer make it a very efficient self-healing material (Supplementary Fig. 2). We made a good trade-off between the mechanical property and self-healing ability by investigating the influence of the ratio between APDS and PPG-TDI on these properties. We proceeded to synthesize three different polymers with APDS to PPG-TDI mass ratios of 11:100, 12:100 and 13:100, which we termed PU11, PU12 and PU13, respectively. The high ratio led to a very soft polymer which could not hold its shape under ambient conditions. At the ratio close to 10.5:100 (1:1 by molar) we could obtain a mechanically strong polymer which exhibits small deformation under mechanical stress (Supplementary Fig. 1). On the other hand, considering the self-healing ability, higher APDS content led to a higher self-healing ability as the obtained polymer is softer (Supplementary Fig. 2). In contrast to the previous material, here we succeeded to obtain a soluble self-healing viscoelastic elastomer, instead of a thermoset, with good mechanical properties, ~1000% strain and ~ 0.26 MPa tensile strength at the break point. For device fabrication, we used PU12 as it showed the best combination between mechanical properties and self-healing ability (Supplementary Fig. 1&3). Similarly to many low $T_g$ polymers/elastomers, PU12 has a very high electrical capacitance (Supplementary Fig. 4). This could be attributed to the high-mobility of chains, the high polarity, and the existence of ionic impurities inside the polymeric structure, such as residues incorporated during synthesis of the elastomer.

Top-contact bottom-gate transistors were fabricated by sequential transfer printing (Fig. 1a). This method is super-convenient with self-healing polymers because of their natural healing ability which enables easy transfer and great attachment of one layer onto another. For the semiconductor preparation, carbon nanotubes (CNTs) were plastered to the surface of the PU12 dielectric in order to obtain both semiconductivity and self-healing ability (Fig. 1c). The same strategy was used to prepare the electrodes. A representative self-healing transistor with a 200 µm channel-length and 5000 µm channel-width was used to evaluate the performance (Fig. 1b). A typical output characteristic of the transistor, showing a good linear and saturated behavior, and a representative transfer curves are shown in Fig. 1d&e. High on-current ($I_{on}$) of ~30 µA, $\mu_h$ of ~10 cm$^2$ V$^{-1}$ s$^{-1}$, and an average on/off ratio of ~$10^3$ were obtained. This is consistent with previously reported CNT transistor using the same semiconducting material. The high
capacitance of PU12 allowed the device to turn on at low gate biases (up to 6 V) which is highly desirable for wearable electronics.

Figure 1  a, Schematic illustration of the fabrication process for the self-healing transistor, and photographs of the self-healing electronic tattoo on skin. b, photographs of a self-healing transistor array on PET flexible substrate (top) and a PI-PS-PI elastic substrate (bottom). c, SEM images of the semiconductive CNT network printed on top of the dielectric layer. Scale bar, 1 µm. d, Output
characteristics of the self-healing CNT transistor with gate voltages from 0 to -4 V in -1 V step, transfer characteristics of the device with drain voltage from -1 to -3. Inset, transfer curve at Vd = -1V (black line), and gate leakage current in the same range (red line).

To prove the autonomic self-healing ability (i.e., self-healing without aiding with any external trigger, such as temperature or light) of each part of the FET, the device was cut using a sharp blade and then followed by monitoring the performance and structural recovery with time. At the beginning, the self-healing ability of the conductive CNT network embedded in PU12 was examined. Figure 2a shows optical images of the healing process of structural damage after making a blade-cut perpendicular to the conduction pathway. This conductive film can recover a drastic cut (> 5 µm in width and ~ 2 µm in depth) quickly and maintains a good conductivity. The self-healing process has a typical behavior with time; a very sharp increase in the conductivity can be seen at the beginning of the recovery, which decreases with time to get a plateau (Fig. 2b). The fast initial self-healing is probably due to shape recovery of the elastic polymer near the damaged area. However, this fast process is followed by slow recovery that might be mainly dominated by the surface rearrangement and diffusion of the polymer chains. After that, the effect of mechanical damage on each part of the FET’s structure and related performance was tested. In the case of gate damage, gate modulation was stopped after the cut, preventing further increase in the drain current (Fig. 2d). The behavior of the FET was almost totally recovered after one hour. The gate electrode could heal several cuts with negligible change in performance (Fig. 2f). On the other hand, the drain current has dramatically decreased (< 10^-10 A) immediately after source/drain (SD) electrode cut and started to increase back towards the initial value. The self-healing process took 1 h and the last value was lower than the original one (Fig. 2e). Several cut cycles in the SD electrode could also be recovered. The initial cut caused the largest decrease in the drain current compared to subsequent damages (Fig. 2g). In contrast to the good recovery obtained with the gate and SD electrodes, similar damages in the channel were too destructive leading to a decreased drain current by almost one order of magnitude after the healing process (Supplementary Fig. 5&6). This significant deterioration in performance is explained by the lower self-healing ability of thinner layers. In the case of CNT semiconductive channel, with a thickness of less than 20 nm, micro-cuts are huge to be totally recovered because the damaged edges are highly deformed preventing the complete reconnection
of CNTs (Supplementary Fig. 7). For real application, we do not expect such big-scale highly-deforming damages because of dynamic attachment.

The efficiency of self-healing was evaluated using four characteristic parameters: $I_{on}$, $\mu_h$, off-current ($I_{off}$), and threshold voltage ($V_{th}$). Starting from the gate damage, the healing efficiency of the four parameters was very high (>98%) (Fig. 2i). However, damages in the SD showed lower self-healing ability. The $\mu_h$ and the $I_{on}$ have decreased by a factor of three while the $I_{off}$ and $V_{th}$ have efficiently recovered to their initial values. Regarding the channel damage, as explained before, the cut was very destructive resulting in low self-healing efficiencies, ~10% with respect to $I_{on}$ and $\mu_h$. The efficiencies of the $I_{off}$ and $V_{th}$ have also been affected but they are still relatively high (>75%). For more severe damage, the whole device was cut through the SWCNT channel by scissors. The performance was monitored after the reconnection of the two parts. Surprisingly, a transistor-like behavior was recovered after ~ 2 hours (supplementary Fig. 8). This was also proved using a LED which was driven by the self-healing transistor; we see in fig. 2h that directly after the reconnection there was no noticeable intensity of light, but it was gradually increased with time.
Figure 2 | The self-healing ability of the transistor. a, Structural monitoring of the self-healing process at different times. Scale bar, 200 µm. b, The dynamicity of the self-healing process in CNT-PU films. c, recovery of several damage cycles in the CNT-PU electrodes. Transfer curves showing the time-dependent healing of the gate (d) and the source drain electrodes (e). Insets show logarithmic scale. f, Transfer curves showing the ability to heal several cuts in the gate. Inset shows a zoom-in on the high current regime. g, Transfer curves showing the ability to heal several cuts in the SD electrodes. Inset shows logarithmic scale. h, photographs displaying the self-healing ability of the transistor after a severe cut. They show the intensity of LED that is driven by the self-healing transistor before cut, directly after reconnecting the two cut parts, and 1 hour after reconnection. i, Self-healing efficiencies of the three different damages based on the four parameters: $V_{th}$, $I_{on}$, $I_{off}$ and $\mu_h$. Efficiency was evaluated according to: $\varepsilon = (f_{Healed} - f_{damaged}) / (f_{original} - f_{damaged})$, where $f$ is the characteristic parameter.
The device performance was tested under uniaxial strain either parallel to or perpendicular to the charge transport direction. First, the devices were conditioned by initial cycles of strain up to 50% rendering them with minimal sensitivity/drain current in subsequent strains. Fig. 3a displays the transfer curves of the transistor under different strain values from 0% to 50%, before and after conditioning. The $I_{\text{off}}$, $I_{\text{on}}$ and $\mu_{\text{h}}$ have decreased by up to ~60% with 50% strain (Fig. 3 c-e). The $V_{\text{th}}$ has slightly shifted to more negative values as the strain increased (Fig. 3 c-f). This might be explained by the small decrease in the capacitance as stretching leads to arranged polymer chains and decreased polarity. After the first stretching cycle, the device became less sensitive to strains where the change in the $I_{\text{off}}$, $I_{\text{on}}$ and mobility with strain is almost negligible. However, the $V_{\text{th}}$ maintains some sensitivity to strain but it is lower compared to the original device (Fig 3c-f). Having both strain-dependent and -independent parameters is desirable for obtaining multi-functional devices that can simultaneously detect strain values and other stimuli such as temperature and humidity without the need for complicated data processing to separate the responses to each type of stimuli. Similar behavior was also observed with uniaxial strains perpendicular to the channel (supplementary Fig. 9). A fatigue test was subsequently performed by applying continuous strain relaxation cycles up to 50% strain. Fig. 3b displays the transfer curves of the transistor after different number of strain cycles. The curves exhibit a negligible change with increasing number of cycles after the conditioning process.
Figure 3] Strain-dependent properties of the self-healing device. a, Typical transfer characteristics ($V_D = -2.0 \text{ V}$) of the device under specific tensile strain applied along the channel length direction, first stretching cycle (solid lines), second stretching cycle (dashed lines). b, Typical transfer characteristics ($V_D = -2.0 \text{ V}$) of the device after cycles of 50% strain. The insets show log-scale characteristics. Normalized electrical characteristics for stretched devices: c, $I_{on}$, d, $I_{off}$, e, mobility and f, $V_{th}$. g, photographs of a device at different strains applied along the channel length direction.

The self-healing transistor is a multifunctional device as it can be used for many applications such as sensing temperature and humidity sensor. The device can be applied on skin as an ultrathin tattoo which is highly promising for future diagnostic applications (fig 4g). The performance of the skin tattoo was tested under different skin strains; the behavior was stable as very negligible changes were observed with different hand positions (fig 4f). Figure 4a display
the responses of the transistor to temperatures between 0° to 60° C. The sensitivity of the sensor in lower temperatures is higher compared to high temperatures; the $I_{on}$ and $I_{off}$ have the biggest changes in lower temperatures (Fig 4b). It might be explained by the fact that lower temperatures limit the polymer mobility/softness which decreases the gate modulation ability and the $I_{on}$ as well. The same effect can be seen on the $V_{th}$ where higher temperatures lead to lower $V_{th}$ values (supplementary fig.10). The increase in the $I_{off}$ with lower temperatures might be explained by the de-swelling of the film leading to a denser carbon nanotube channel. This sensing ability was conserved after the healing of a structural damage. Fig. 4b compares the normalized responses of pristine and recovered sensors. In this case, the mechanical damage affected all parts of the transistor (supplementary Fig. 11). It is clearly seen that the difference between the two cases is negligible. A self-healing wearable skin hydration sensor was also demonstrated. PU12 is highly affected by humidity, as water molecules can be absorbed or emitted leading to changes in the elasticity of the materials and the capacitance as well. This change can give a good indication for the hydration levels of the skin. In this experiment, lotion was applied on the skin to increase its hydration level, and then skin humidity was monitored by two sensors, the self-healing epidermal sensor and a commercial skin humidity sensor. The $I_{on}$ and the $V_{th}$ were used to monitor the effect of humidity levels. Fig. 4c displays the transfer curves obtained different times after the application of lotion on the skin. The transistor shows lower $V_{th}$ directly after the humidification which goes and decrease slowly with time approaching the original value. The same trend was seen with the $I_{on}$. Fig. 4b displays the results of this experiment; the same trend was obtained in both sensors. In addition, the same behavior was seen after the self-healing of the sensor.

To summarize, we have introduced the first autonomic self-healing and highly stretchable FET using a self-healing PU as a host matrix filled with conductive or semi-conductive CNTs. The self-healing transistor is able to recover several and varying mechanical damages ranging from micro-cracks to complete cut of the whole device by scissors. Moreover, the device can be repeatedly stretched for more than 200 cycles of up to 50% strain without a significant loss in electrical properties. The self-healing and high stretchability of the device improves its lifetime as well as potentially enhance its autonomic functionality. The transistor can be used as a sensor which detects temperature and humidity. In addition, the self-healing device was applied as an
ultrathin temporary tattoo on skin. This platform is very promising for in situ analysis of skin chemical and physical environments by modern epidermal applications that facilitate personalized and continuous physiological and clinical investigations.

**Figure 4** Transistor-based self-healing applications. 

**a**, Transistor sensitivity to different temperature: Responses of the $I_{on}$ (black line) and $I_{off}$ (blue line). 

**b**, Normalized responses of $I_{on}$ and $I_{off}$ before and after self-healing. Similar responses are obtained in both cases indicating that the self-healing ability of the sensor is very efficient. 

**c**, Transfer curves obtained from the humidity epidermal sensor different times after skin humidification. 

**d**, Normalized responses of the $I_{on}$ and $V_{th}$ in comparison to the normalized response of a commercial sensor before and after self-healing. The damage has a negligible effect on the sensitivity of sensors that completed the healing process. 

**e**, photographs of the epidermal humidity sensor mounted on the skin of the arm. 

**f**, performance of the self-healing transistor skin tattoo in different hand positions that leads to different strains on the skin tattoo. 

**g**, photographs of the epidermal self-healing skin tattoo during measurements.
Methods

Materials. All solvents were received from commercial sources, and used as received. Poly(propylene glycol), tolylene 2,4-diisocyanate terminated (PPG-TDI), glycerol, 4-aminophenyl disulfide (APDS), Triton X-100, Polyvinyl alcohol (PVA), and polystyrene-block-polyisoprene-block-polystyrene (PS–b-PI–b-PS, 17% styrene) were obtained from Sigma-Aldrich. CNTs for electrodes (>75%) were purchased from TUBALL. Semiconducting CNT aqueous solution (+99%, 0.01 mg/ml) was obtained from Nanointegris. FluorN 561 flurosurfactant was obtained from Cytonix.

Device Fabrication. PS–b-PI–b-PS, which is stretchable and mechanically durable and most importantly, it also exhibits some kind of a self-healing ability as shown in a recent study. Substrates were prepared by dissolving 80 mg of PS–b-PI–b-PS in 1 ml of Toluene, and casting into a Teflon mold. When dried in hood, the sample was left under vacuum to obtain ~500 µm thick elastic substrate. Solutions for CNT electrodes were prepared as by dispersing 1 mg of CNTs in 1ml water using 5 mg/ml Triton X-100 as a surfactant. The solution was centrifuged at 6000 rpm for 30 min before use. For electrodes preparation, CNT dispersion was sprayed through a shadow mask on a slightly modified silicon wafer which was prepared by treatment with oxygen plasma and then immersion in toluene containing Octadecyltrichlorosilane (ODTS) for 1 min only. Long immersion time leads to very hydrophobic surface which is not convenient for spraying the aqueous dispersion of CNT. On the other hand, PU is very sticky to the non-modified wafer preventing subsequent peeling off. 200 mg/ml PU precursor solution in THF consisting of PPG-TDI and APDS with a mass ratio of 100:12 were drop casted on the gate electrode, left to dry for 1 min and followed by spinning at 1000 rpm for 1 min to obtain ~20 µm PU substrate. Then it was transferred to substrate or PS-b-PI-b-PS elastic film by applying gentle pressure and heating. The same solution was spin-coated on the source drain electrodes at 2000 rpm to from ~2 µm thick dielectric, which was transferred later onto the gate electrode by applying gentle pressure and heat. SWCNT ink was prepared by mixing 99% Semiconducting-SWCNT solution, DI-water, propylene glycol and FluroN at the ratio of 1:6:1.25:0.75 (by volume), followed by ultrasonication. Subsequently, a 30 µL of SWCNT ink was drop-casted on the CNT source/drain composite electrodes and heated to 90ºC for 30min. The device was rinsed.
in DI water for 1 hr to remove the surfactants, and then left in vacuum oven under 90° C for several hours. It is worth mentioning that the whole device can be potentially printed since all materials are solutions-processable, enabling high-throughput production.

**Preparation of self-healing skin tattoo.** The same method, as described above, was used but instead of transferring the device to PS-PI-PS, it was transferred to a PVA-coated paper. In addition, the device was coated with another layer 1 µm PS-PI-PS and 1 µm PDMS (on top of the channel and SD electrodes) to prevent direct contact with the skin. The printed tattoo was applied on skin by gentle press and then washed with water to dissolve the PVA layer and release the tattoo to the skin.

**Electrical and Mechanical Characterization.** The dielectric constant and capacitance of the elastomeric dielectric were measured using LCR meter. Two-point resistance measurements were done on the CNT electrodes using a Keithley (model 2701 DMM). Transistor characteristics were collected using a Keithley (2636A SYSTEM SourceMeter) controlled by a custom Labview program, in ambient atmosphere. Devices were conditioned in ambient before measurements in order to minimize the confounding effects of water absorption in the dielectric. Stretching for electrical performance was accomplished using a manual apparatus. Mechanical stress-strain experiments were performed using Instron to evaluate the tear strength and tensile properties of the polymers. The stretching rate was 100 mm min⁻¹.

**Self-healing tests.** The self-healing processes were monitored using an optical microscope (BX51M, Olumpus) with integrated camera (LC20, Olympus). SEM was also used to detect the self-healing of the semiconductive channel. Electrical self-healing characterization of electrodes was done by two-point resistance using a Keithley data logger device (model 2701 DMM), controlled by a custom Labview program which was used to acquire sequential resistance readings from CNT conductive films. Cutting was done with a ~20 µm blade or >1 µm razor-sharp blade. Self-healing characterization of the transistor was done by measuring the device performance before damage, while damaging the device, and different times after that. Four different damages were done: blade cut through the source/drain, gate, and the semiconducting channel, and cutting the whole device by scissors and then reconnecting both parts.
**Sensing Experiments.** For temperature sensing, the transistor was monitored while it is attached to a surface with fixed temperatures ranging from 0° to 60° C. The transistor was covered with PS–b-PI–b-PS and PDMS to prevent the effect the external environment (e.g. humidity). For skin humidity, lotion was applied on the skin to increase its hydration level, and then skin humidity was monitored by two sensors, the self-healing sensor and a commercial digital moisture monitor for skin (model SK-IV). In this experiment, the transistor was covered with PDMS to prevent direct attachment to the skin. For these two experiments, the damage that we made affected all components of the transistor, as explained in supplementary fig. 11.
Figure S1. Stress-strain measurements. a, Images of the system while stretching the self-healing polymer. b, Stress-strain behavior of the three self-healing polymers before cut (solid lines) and after 24 hr of healing (dashed lines).
Figure S2. DSC curve recorded for PU12 with a scan rate of 10 °C min⁻¹.
Figure S3. Self-healing process of small and big cuts made by sharp blades on the three polymers. A, PU13, B, PU12, and C, PU11. Scale bar 200 μm.
Figure S4. Capacitance properties of PU12. a, Capacitance over frequencies ranging from 0.1 to $10^6$. b, Capacitance as a function of strain.
**Figure S5.** Transfer curves showing the time-dependent healing of the semiconductive channel. Inset shows logaretmic scale.

**Figure S6.** Optical images of the recovery of surface scratches; immediately after scratch (left), 30 min after scratch (middle), and one day after (right).
Figure S7. Scanning Electron Microscope (SEM) images of a damaged area after partial recovery. Circled area presents a well-recovered part where the CNTs from both cut-sides are reconnected.
Figure S8. Recovery after cutting the whole device in two parts through the channel. Immediately after reconnection, the device shows positive drain currents which arises from gate leakages, while after one day a transistor-like behavior was returned although the healing efficiency if very low. Very dense channel was used in this experiment which led to the low on/off ratio.
Figure S9. Device stretchability perpendicular to the channel conduction pathway. a, Typical transfer characteristics (V_d = -2.0 V) of a the device under specific tensile strain applied perpendicular to the channel length direction, first stretching cycle (solid lines), second stretching cycle (dashed lines). b, typical transfer characteristics (V_d = -2.0 V) of a the device after cycles of 50% strain.
Figure S10. Sensitivity of the $V_{th}$ to temperature.
Figure S11. Illustration image of the structural damage made in the self-healing experiments.

Figure S12. Hysteresis characteristic of the CNT self-healing transistor at 0.02 V/s gate voltage sweep rate.