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Electrochemical circuits from 'cut and stick' PEDOT:PSSnanocellulose composite

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Abstract

We report a flexible self-standing adhesive composite made from PEDOT:PSS and nanofibrillated cellulose. The material exhibits good combined mechanical and electrical characteristics (an elastic modulus of 4.4 MPa, and an electrical conductivity of 30 S cm⁻¹). The inherent self-adhesiveness of the material enables it to be laminated and delaminated repeatedly to form and reconfigure devices and circuits. This modular property opens the door for a plethora of applications where reconfigurability and ease-of-manufacturing are of prime importance. We also demonstrate a paper composite with ionic conductivity and combine the two materials to construct electrochemical devices, namely transistors, capacitors and diodes with high values of transconductance, charge storage capacity and current rectification. We have further used these devices to construct digital circuits such as NOT, NAND and NOR logic.

1. Introduction

The most explored and studied organic electronic material to date is poly(3,4-ethylenedioxythiophene): poly(styrene-sulfonate) (PEDOT:PSS). Originally developed as an antistatic agent coating [1], it is nowadays used in a number of applications, such as the charge-injecting electrode in electroluminescent devices [2], photovoltaics, and field effect transistors, and as the active channel or pixel in electrochemical transistors and electrochromic displays [3]. The work function of PEDOT, while in its highly conducting state via charge compensation provided by a highlycharged molecule, is around 5.1 eV and PEDOT is therefore often used as a hole-injection electrode material in the field of printed organic electronics [4]. PEDOT:PSS is commonly sold as an aqueous dispersion, and ready-made PEDOT:PSS films are available on a variety of substrates (plastic foil, cardboard, etc) and may be obtained from multiple commercial vendors.

In early 2000, it was discovered that the addition of high boiling point solvents could further boost the electrical conductivity of PEDOT:PSS by three orders of magnitude, making PEDOT the first air-stable, water-processable and actually useful plastic conductor [5]. This opened up completely new avenues for PEDOT:PSS and made it successful in a vast array of applications. Some of the first efforts towards applications aimed at electronic conductors include electrodes in transistors [6], flexible electrodes [7], transparent electrodes in OLEDs and OPVCs [8], as well as thermoelectric materials [9]. PEDOT was also explored in various electrochemical devices. Indeed, the PEDOT:PSS charge compensating composite transports both electronic charge carriers (PEDOT phase) and ionic charge carriers (PSS phase); PEDOT: PSS is a mixed ion-electron conductor. The ability to conduct both types of charge carriers enables its use in electrochemical devices, where it can serve as both the active material and the electrode. Via electrochemical switching, one can introduce modulation in (i)

electronic conductivity which is utilized in electrochemical transistors [10]; (ii) its color then explored in electrochromic displays [11]; and (iii) ion–electron charge compensation being used in supercapacitors [12]. The multifunction of a single material makes it very attractive to simplify the manufacturing process of complex printed electronic systems, since several different devices can be made out of the same thinfilm material [13]. However, all the devices based on, or including, PEDOT:PSS listed above are actually a thin-film technology manufactured on a relatively much thicker substrate.

Here, we report and demonstrate electrochemical devices and circuits, based on a high performing PEDOT:PSS-nanocellulose composite, which is possible to assemble and reconstruct using a self-adhesive assembly and reconfiguration approach. Our work is motivated by the fact that in many applications, it is desirable for the functionality to be included in the substrate. For instance, several devices, such as charge storage devices and power electronics, require large currents and are therefore thick. Adding those onto flexible substrates typically introduces undesired volume (the substrate) to the system, whereas the approach proposed here integrates the devices into the substrate itself. Moreover, with an electroactive carrier, new simplified routes towards double-sided printed electronics arise. In addition, with self-adhesive electronic substrates, new manufacturing protocols based on cut-and-stick approaches make label-assembly of devices and circuits possible. Finally, such substrate-free electronics would also enable new types of electronic devices [14], where the chemical and physical properties of the paper bulk are now possible to control via electronic addressing. In that spirit, Lee et al reported a 'cut and stick' ion gel material used as the dielectric in low-voltage organic and inorganic thin-film transistors [15]. These devices, however, rely on metal electrodes (atop a substrate) for the transistors' terminals. Moreover, the active material is not part of the 'cut and stick' gel. Generally though, the field of organic and printed electronics has been limited to the coating of thin films onto substrates such as plastic and paper [16].

Nanofibrillated cellulose (NFC) is a nano-structured form of the most abundant natural polymer on the planet, i.e. cellulose [17]. The cellulose is converted into fibers via chemical treatments and is thereafter delaminated into nanofibrils using high-pressure homogenization [18]. Thanks to the resulting narrow diameter, NFC has a high specific surface area. NFC can be processed into various kinds of paper- and plastic-like substrates from solution thanks to the fibrils' large aspect ratio (>1000) [19] and colloidal stability. NFC films has been shown to exhibit superior tensile strength and elastic modulus compared to their fiber counterpart [20].

Kawahara *et al* built upon the 'cut and stick' concept and developed self-adhesive, reconfigurable sticker label electronics based on NFC and PEDOT: PSS [21]. They demonstrated how freestanding conductive films could be laminated to form electrochemical devices such as organic electrochemical transistors (OECTs) [3]. However, only the gate of the transistors was comprised of the electronic composite films while a commercial plastic with a thin film coating of PEDOT:PSS was used as the channel. Another combination of NFC and PEDOT:PSS was subsequently reported and demonstrated '3D' (thick film) organic power electronic devices [14]. Although not self-adhesive, the latter material was used to construct a paper-based OECT with a record high transconductance of 1 S operating at current levels over 1 A.

Here, we combine the concepts of 3D organic power electronics and self-adhesive reconfigurable materials based on cellulose nanofibrils, PEDOT:PSS and polyelectrolytes in order to demonstrate a variety of electrochemical devices and circuits.

2. Results and discussion

2.1. Material fabrication

To fabricate the (semi)conducting composite material, we used a blend of PEDOT:PSS solution (Clevios PH1000), NFC (aqueous solution at 0.42 wt% concentration), glycerol and dimethyl sulfoxide (DMSO) in the following weight percentages: 6.5/2.5/27.7/63.3. Figure 1 shows the molecular structures of the different materials. The solution is dispersed with a high-shear batch mixer for 2-3 min and then solvent cast into plastic Petri dishes and dried at room temperature [22]. NFC has a high toughness with a large strain-to-failure [20] and serves as a mechanically robust scaffold onto which the conductive polymer self-organizes [14]. The addition of glycerol is dualpurposed: it works as a plasticizer and it improves the films' hygroscopicity (because of glycerol's three hydroxyl groups) [23]. DMSO acts as a secondary dopant by stretching the PEDOT coils, therefore increasing the interchain interactions [24]. The resulting material conforms easily, sticks to most common surfaces, and is very resilient to deformations. The flexibility of the films is demonstrated in figure 1 where a $\sim 2 \times 2$ cm sheet is rolled up.

2.2. Electrical properties

Figure 1 displays a slice of a thick (6 mm) piece of the material. To determine how the electrical properties are affected when going from thin to thick films, the conductivity was measured for various thicknesses. The results (figure 2(a)) show that the conductivity remains relatively constant over a wide range of thicknesses (30–600 μ m) with an average conductivity of 30 S cm⁻¹, indicated by the dashed line. This suggests that the composited material is homogeneous from the scale of about 10 μ m. When normalizing the electronic conductivity with the volume fraction of



Figure 1. Photographs of a 6 mm thick slice and a rolled-up film of the conductive composite, and the molecular structures of its components.



Figure 2. (a) Conductivity versus thickness (n = 4), (b) conductivity (n = 4) and elastic modulus (n = 4) versus weight percent of nanofibrillated cellulose (lines are b-spline interpolated), (c) resistance vs sample length and (d) resistance versus vertically stacked layers (length 10 mm). The dashed line in (a) shows the average of the mean values, the dashed lines in (b) are meant as guides for the eyes and the dashed lines in (c) and (d) are linear fits.

PEDOT:PSS of the bulk the conductivity reaches $386 \,\mathrm{S}\,\mathrm{cm}^{-1}$, which puts it in the range typically reported for PEDOT:PSS [25].

The electrical conductivity, as well as the tensile stress and strain, was further investigated for samples

with different amounts of NFC. The stress and strain data were used to calculate the elastic modulus, which can be seen together with the conductivity in figure 2(b). As expected, there is a tradeoff between the elastic modulus (which represents the stiffness) and the conductivity since NFC is an electrical insulator. The amount of NFC in our standard recipe is 2.5 wt%. From the data reported in the graph of figure 2(b), even such a small amount has a large impact on the elastic modulus as compared to samples without NFC. With only 2.5 wt% NFC, the composite's resulting elastic modulus increases by a factor 5. When adding 20 wt% of NFC, the elastic modulus increases by more than 300 times. However, at this point, the conductivity also drops significantly. To avoid compromising the electrical conductivity, the NFC content was kept low. An additional reason for doing so is to preserve the stretchability of the material. As can be seen in figure SI1 available online at stacks.iop.org/ FPE/2/045010/mmedia in the supplementary information, although the tensile strength (stress at break) increases with the amount of NFC, strain at break decreases, meaning that the material becomes less stretchable.

Although the elastic modulus in the standard recipe (4.4 MPa) is small as compared to that of paper, which is typically in the GPa-range [20] this is still a relatively large value considering that the solid part, represented by PEDOT:PSS and NFC, only makes up 9 wt% of the resulting nanopaper, making it more a gel rather than a solid. The large span in the elastic modulus makes it possible to tailor the material's mechanical properties by changing the NFC content. In this way, the elastic modulus can be matched to that of the substrate onto which the nanopaper is adhered. The elastic modulus of human skin is in the range 0.05-20 MPa, as the reported values vary with the measurement techniques [26]. This range partly overlaps with the range of the nanopaper, enabling it to be used for applications such as electronic skin [27] or transdermal drug delivery systems [28].

Since the material is meant to be stacked in different configurations to form electrochemical devices, one important parameter is contact resistance. Figure 2(c) shows the resistance of a nanopaper sample contacted with gold-coated plastic (PET) sheets with different spacing between the contacts. The inset of the figure shows how the sample was contacted as well as the dimensions. For each spacing, the sample was laminated and delaminated five times (represented by the error bars in the graph). The error varies between 10 and 100 m Ω , indicating that the differences in pressure during lamination have very little impact on the characteristics of the electrical contact. The material conforms to smooth surfaces without any applied force. In fact, applying force does not improve the electrical contact, as can be seen in figure SI2 in the supplementary information. Using a linear fit, we find that the intersection with the y-axis in figure 2(c)occurs at the contact resistance of 4 Ω (2 Ω for each contact). However, this value coincides with lateral resistance of the gold foil used, which in fact suggests that the contact resistance is smaller than the accuracy of the measurement ($<100 \text{ m}\Omega$).

A similar approach as in figure 2(c) was used to measure the contact resistance between two nanopaper films when laminated. The resistance was measured as additional layers of nanopapers were added between the gold contacts and the first original nanopaper (in this case with 10 mm separation between contacts). Figure 2(d) shows the resistance for different number of layers and the inset shows the measurement setup. As with the previously mentioned measurement, the sample was laminated and delaminated repeatedly (three times) for each number of layers. The variation in the overall resistance in between the lamination steps was found to be in the range from 30–170 m Ω , which was similar to the results in the previous measurement (see above). By linear fitting, the average increase in resistance for each layer was determined to be 85 m Ω . The resistance is a combination of contact resistance between the layers and the vertical bulk resistance. This sets an upper limit for the contact resistance to 85 m Ω for an area of 25 mm² (or 2.13 Ω mm²) and a lower limit to the vertical conductivity of the bulk to $\sim 600 \text{ mS cm}^{-1}$. This means that there is a relatively small difference between the in-plane (σ_{IP}) and out-of-plane (σ_{OP}) conductivity $(\sigma_{\rm IP}/\sigma_{\rm OP} < 50)$. This is in great contrast to spincoated thin films of PEDOT:PSS where the ratio between the in-plane and out-of-plane conductivity can be up to 30 000 for certain polymer blends [29].

2.3. Adhesive properties

In addition to the electrical properties of lamination of the conductive films, the mechanical properties of the contact between films were also investigated. One desired characteristic of a reconfigurable 'sticky label' is that it should exhibit strong cohesive forces, to ensure the integrity of the label itself, high enough adhesive forces to promote stable adhesion to hosting surfaces and at the same time low enough adhesive forces to enable easy peeling off. To quantify the forces needed for peeling and 'pulling' the nanopapers apart, the setups shown in figures 3(a) and (b) were used. Figure 3(a) shows a photo of the apparatus used to separate samples and simultaneously measure the force, as well as the sample setup used to measure the force of peeling. A different set of contacts on the apparatus was used for the pulling measurements. A schematic picture of the pulling setup is depicted in figure 3(b) where cellophane film was glued on the metal contacts with the nanopaper sandwiched in between.

Figure 3(c) shows the results of the pulling measurement. The measurement was repeated 4 times for the same sample and an average peak force of 5 N was recorded. Figure 3(d) shows the result of the peeling measurement. Here, the measurement was performed once for two different samples with an average maximum force of 11.5 mN. The sample used for pulling was circular with an area of 10 cm² while the samples



used for peeling were 2 \times 6.5 cm (13 cm²). The force needed to peel the sample does not depend on the total area of the sample, but rather the width (the dimension perpendicular to the direction of peeling). This is because only the interface between the contacted areas and the part of the sample which has been separated contribute to the force. This is evident from the relatively constant force in the peeling measurement which should instead decrease linearly if the force was determined by the total contacted area. However, a wider sample will have a larger contact at the interface and will therefore require a larger force to separate, making the width of the sample the force-defining parameter. The force in the pulling measurement, on the other hand, is proportional to the contact area of the sample since the force is spread homogeneously during the measurement. Therefore, the force needed to pull a sample of the same dimensions as the sample used for peeling $(2 \times 6.5 \text{ cm})$ would be 6.5 N. The maximum force needed to pull such a sample would, therefore, be 565 times larger than for peeling it. This result shows that the nanopapers work well as reconfigurable adhesives, since the large difference in force means that the papers will stay in place once laminated, but can still be removed without breaking them if they are peeled off.

The self-adhesive properties of the nanopaper in combination with its electrical functionality and mechanical robustness make it an ideal material for electrical contacting to rough, soft and even fragile surfaces. The material has been used to interface with biological systems such as plants [30] and could be extended to human skin for EEG/EKG and skin conductance measurements. For such applications, DMSO (which can penetrate the skin) could be replaced with a more biocompatible solvent like ethylene glycol. The softness of the nanopaper also enables it to be used for probing sensitive systems, which would otherwise be damaged by attaching metal contacts. After the measurement, the contacts could be removed without damaging the sample and then reused.

2.4. Electrochemical devices

An ionically conductive paper was prepared by soaking cellulose filter papers into a mixture of a quaternized polycation (Luviquat Excellence) and glycerol. This electrolyte paper, together with the conductive nanopaper were used to construct different electrochemical devices, namely, an OECT [3], a pseudocapacitor [14, 31] and an electrochemical diode [32]. Figure 4(a) shows a schematic illustration of the layers of each device. In the case of the OECT, two gate electrodes (instead of one) and two electrolyte layers were used to achieve a better on/off ratio. The two gates connect to each other forming a 'wrap-around gate'. Figure 5 also shows photos of the electrolyte paper (figure 5(a)) and the transistor (figure 5(b)).

When a voltage is applied between the transistors' drain and gate (with the negative potential on the



drain), the PEDOT:PSS undergoes an electrochemical reaction. The gate is oxidized while the channel is reduced. In the reduced state, the PEDOT phase, and thus the channel, has a low electronic conductivity, which turns the transistor off. The transfer characteristics of the OECT can be seen in figure 4(b) showing the drain current and the transconductance versus gate voltage. The drain voltage was kept at -1.25 V while the gate voltage was swept from 0 to 2 V. The transistor exhibits an on/off ratio of 237 at $V_{\rm G} = 1.7 \,\rm V$ and a maximum current of 45 mA at $V_{\rm G} = 0$ V. The maximum transconductance reaches 45 mS at $V_{\rm G} = 0.6$ V, a value comparable to the stateof-the-art [10]. The output characteristics and switching measurements of the transistor can be found in supplementary information (figures SI3 and SI4).

A pseudocapacitor was constructed from two pieces of nanopaper $(1 \times 1 \text{ cm})$ separated by an electrolyte paper. When a voltage is applied between the electrodes, the PEDOT:PSS in the positive electrode becomes oxidized while the negative electrode is reduced. Ions migrate in and out between the nanopaper electrodes and the electrolyte paper in accordance with charge conservation. Figure 4(c) shows cyclic voltammograms of the pseudocapacitor at scan rates of 25, 50 and 100 mV s⁻¹. The current versus voltage characteristics for the measurement ran at 25 mV s⁻¹ shows the most 'box-like' behavior, characteristic for a capacitor. At higher scan rates the device performance becomes limited by the slow ion migration in the electrolyte and within the nanopaper electrodes. The capacitance at this scan rate was calculated to be 20 mF cm⁻² using the formula C = I/(vA) where *I* is the current a 0 V, *v* is the scan speed and *A* is the area.

An electrochemical diode was constructed using a large electrode for the negative end of the diode and a smaller stripe for the positive end. The two pieces of nanopaper were partly separated by an electrolyte paper, but in contrast to the pseudocapacitor, they were also in electronic contact. When a voltage is applied across the diode there will be an electronic current running through the device. However, due to



the potential difference between the two terminals, there will also be an ionic current going through the electrolyte paper. This induces a similar electrochemical reaction as in the transistor and the pseudocapacitor; i.e. the nanopaper on the positive side of the electrolyte will be oxidized while the other side becomes reduced. In forward bias, the top electrode becomes oxidized while the bottom one becomes reduced. However, only part of the bottom piece is in contact with the electrolyte paper, and so the resistance between the terminals will be mostly unaffected. At reverse bias, the top electrode becomes reduced and its conductivity decreases. Since the full width of this electrode is in contact with the electrolyte, the total resistance of the device will increase and the current will decrease. Figure 4(d) shows a voltage sweep of the diode from -3 V to +3 V at a scan speed of 2 mV s⁻¹. The rectification ratio at ± 3 V reaches 10, one order of magnitude smaller than previous reports [32].

The performance of the nanopaper electrochemical devices is vastly different to the reported performance of their thin-film counterparts. The main difference is that considerably larger currents can be passed though the nanopaper-based devices, although they respond much slower. The current passed through the nanopaper OECT is more than 5000 times larger than the previously reported OECT based on the cut and stick method which used a thin film of PEDOT:PSS (200 nm) coated on a plastic substrate as the channel [31]. However, the on/off ratio of the reported device was in the same order (~100) as for nanopaper OECTs, even though the thickness of the channels differed by several orders of magnitude. The on/off ratio is still small as compared to other reported OECTs, with values often reaching as high as 10^5 [3]. This property of the transistor is largely dependent on device dimensions and the electrolyte, and further optimization of the nanopaper OECT might improve its performance.

Both the large current density and long response time is attributed to the thickness of the devices and not to the material itself. This class of devices is therefore suitable in applications where large conductance and charge storage is preferred over a fast operation.

2.5. Electrochemical circuits

The 'cut- and stick'-OECTs were further combined into circuits in order to realize logic NOT, NAND and NOR gates. These logic circuits were previously realized using thin-film OECTs [10]. Figure 5(c) shows a picture of one such circuit (NAND) and figure 5(d) show schematics of each circuit layout. A set of three variable resistors were constructed by stencil printing lines of PEDOT:PSS on a plastic substrate using a barcoater. Each pair of neighboring lines was then connected by a stripe of nanopaper to form a resistor.



By moving the stripe along the lines, the resistance between the endpoints of each line may be varied. Each resistor was subsequently connected by additional nanopaper stripes. The electrical characteristics of the three logic gates can be seen in figure 6. The time to switch between two states was around 30 s.

3. Conclusions

In conclusion, we report a scalable manufacturing method to produce flexible self-standing functionalized nanopaper composites. These films exhibit excellent mechanical and electrical properties as well as good self-adhesive and re-adhesive characteristics, properties that can easily be tailored to different applications by adjusting the material recipe. We demonstrated how these materials can be used to construct electrical and electrochemical components and circuits by a 'cut and stick' assembly method. Since the films are self-standing, this manufacturing method could nicely complement already established printed electronics techniques [33]. The 'cut and stick' aspect of this material naturally lends itself to reconfigurability and recycling of the building blocks, and devices could be imagined to be assembled using pick and place machines or by end-users that reconfigure system functionality by simply reorganizing the labels. Batteries and supercapacitors are examples of devices that might benefit from thick and high capacity electrodes. The high current-carrying ability enabled by this composite material opens up the door for applications within the field of organic power electronics [14]. Finally, the combination of good electrical (high conductivity and small contact resistance), mechanical and adhesive properties makes it an attractive material to be used as contacting electrodes to electronic or biological systems with rough and/or sensitive surfaces.

4. Experimental section

4.1. Materials

PEDOT:PSS solution, Clevios PH 1000, was purchased from Heraeus. Glycerol, dimethyl sulfoxide (DMSO) and polystyrene sulfonic acid (PSSH) were purchased from Sigma-Aldrich. Poly(1-vinyl-3methylimidazolium-co-vinylpyrrolidone)-chloride (Luviquat Excellence) was purchased from BASF. Nanofibrillated cellulose (NFC) was produced by carboxymethylation of cellulose dispersed in water (degree of substitution of 0.1) followed by ten highpressure (1700 bar) microfluidization iterations. The resulting concentration of the NFC dispersion was 0.42 wt%.

4.2. Material fabrication

Clevios PH 1000, NFC solution, glycerol, and DMSO were mixed in a solid content (excluding only water) weight ratio of 6.5/2.5/27.7/63.3. The solution was mixed using a T 10 basic ULTRA-TURRAX (Laboratory Mixers) for several minutes followed by degassing for 1 h in a vacuum desiccator. The solution was then cast in plastic Petri dishes and left to dry. Films with different thicknesses were manufactured by adding different volumes of solution to the Petri dishes. The standard thickness of the samples used for electrical and mechanical characterizations, as well as for the devices and circuits, was 120 μ m (±10 μ m), unless otherwise stated.

The electrolyte paper was prepared by mixing Luviquat Excellence (40 wt% in water) with anhydrous glycerol with the mass ratio 5:4. The solution was stirred for 1 h after which 1.5 ml was soaked into a filter paper with the area 23 cm². The paper was left for >4 h before being used to let excess water evaporate.

4.3. Electrical characterization

A plastic foil was coated with 6 nm titanium followed by 60 nm gold using thermal evaporation. The foil was then used to contact the conductive paper during electrical measurements.

Contact resistance measurements and conductivity measurements were performed using a Keithley 2400 source meter. The contact resistance measurements were performed in a two-wire measurement mode while the conductivity measurements were performed in a four-wire measurement mode.

The transistor measurements were performed on a Keithley 4200. The measurements on logic gates were performed using two Keithley 2600.

4.4. Mechanical characterization

All mechanical tests were performed at room temperature of 23C and 50%RH and at a pulling speed of 100 mm min⁻¹. Tensile stress–strain measurements were performed for samples with varying amount of NFC with length 30 mm and width 6 mm. 4 samples were used for each NFC concentration. Peeling measurements were performed by laminating two samples with dimensions 2×6.5 cm followed by pulling them apart as demonstrated in figure 3(a). The samples were left for 20 min between lamination and measurements. Pulling measurements were performed by sandwiching a sample with the area 10 cm^2 between two cellophane membranes which had been glued to metal contacts (depicted in figure 3(b)). The force needed to pull apart the metal contacts were then measured.

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