Green and biodegradable electronics

We live in a world where the lifetime of electronics is becoming shorter, now approaching an average of several months. This poses a growing ecological problem. This brief review will present some of the initial steps taken to address the issue of electronic waste with biodegradable organic electronic materials. Many organic materials have been shown to be biodegradable, safe, and nontoxic, including compounds of natural origin. Additionally, the unique features of such organic materials suggest they will be useful in biofunctional electronics; demonstrating functions that would be inaccessible for traditional inorganic compounds. Such materials may lead to fully biodegradable and even biocompatible/biometabolizable electronics for many low-cost applications. This review highlights recent progress in these classes of material, covering substrates and insulators, semiconductors, and finally conductors.

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Sustainability is "the ability of a generation to ensure its needs for the present without compromising the ability of the future generations to meet their own needs"[1-3]. While it is difficult to provide a thorough description for consumption, the definition provided in reference 3 seems to incorporate the multitude of problems that consumption poses to the sustainable development of modern society: "Consumption is the human transformation of materials and energy (along the production-consumption chain) that makes the transformed materials or energy less available for future use, or negatively impact biophysical systems in such a way to threaten human health, welfare, or other things people value".

Plastic consumption and waste are two of the major concerns in the modern world. Polyethylene for example is currently the leading plastic material, with a global consumption of about 83 million metric tons in 2010, mostly for use in plastic bag production (>1 trillion/year)[4,5]. Due to the increased demand in countries with emerging economies, plastics consumption is projected to increase approximately by a factor of three during the current decade[6]. The outcome of the constant demand for plastics is the buildup of non-biodegradable solid waste and plastic litter (estimated at 25 million tons/year in the year 2000) with negative consequences on our environment[7,8]. Plastics normally biodegrade very slowly, with full degradation occuring after 500 or 1000 years[9,10].

ISSN:1369 7021

Plastic electronics on the other hand represent an emerging field of science and technology that began in the realm of academic curiosity thirty years ago, but which has now made some amazing advancements. The industrial and commercial potential of organic molecules has been demonstrated in recent years through mature OLED technology and the recent surge of organic photovoltaics (OPV), with reports of efficiencies of about 10 % or higher in 2012[11-13]. Samsung produced 45 million OLED displays in 2011 and projects to build up to 600 million units by 2015[14], whereas printed flexible photovoltaics are currently commercialized at a smaller scale for rooftop and small appliance applications. Although compared to the global production and consumption of plastics in the world, the organic electronics market contributes only a small amount of waste, this number should not be neglected. A symbolic example of the problems that e-waste poses to the environment is presented in Fig. 1. In the race to match the performance of inorganic materials and realize attractive new consumer products like OLED displays, the issues of biodegradability and biocompatibility of the materials employed in organic electronics are often not considered. Up to the present, a large amount of research has focused on synthetic avenues for the production of active layers in organic field effect transistors (OFETs), organic light emitting diodes (OLEDs), and organic photovoltaics (OPVs) – neglecting how these materials affect the environment, as well as questions of toxicity to humans, animals, and plants. Nevertheless, the true biocompatibility of such materials is of paramount importance, not only for the development of biomedical devices and applications involving interfacing with living tissue[15-17] but also for *human friendly* electronics in general[18,19]. The development of neural prosthetics, neural implants, drug delivery devices, and diagnostic electronics all require materials that are the least invasive[20-24]. Research in this area is relatively well-developed compared to 'green' technology for organic optoelectronics. This success can be explained by considering the unique advantages of organic conducting materials: (1) the combination of mechanical robustness with flexibility; (2) nontoxicity and the property of not eliciting inflammatory or immune responses; and (3) the ability to behave as ionic and electronic conductors, and thus interface electronics with the protonic and ionic currents present in biological systems. This brief review will present the recent advancements in terms of biocompatible/biodegradable materials as well as technologies and devices. In the following, organic materials will be classified according to their functionality: substrates and insulators, semiconductors, and conductors.

Substrates and insulators

Numerous materials with a bio-origin have been identified as suitable substrates for the fabrication of organic electronics. Additionally, many of these materials demonstrate excellent insulating properties, which combined with the ease of their processability make them suitable as gate electrode insulators for OFET applications. Such materials enable several functionalities: low-cost, non-toxicity, biodegradability, and often biocompatibility and bioresorbability for biomedical applications.

One of the oldest and most familiar 'substrate' materials of natural origin is paper. Paper is made from plant-derived cellulose. Many varieties of paper are known, and the science of mass-producing paper with desirable mechanical and surface properties is mature. It is by far the cheapest biodegradable substrate material and enables large-area printing of 'use-and-throw' devices. Arrays of OFETs and OFET circuits have been printed on paper, demonstrating flexible devices with performance on-par with more traditional substrates (Fig 2a)[25,26]. Low-voltage active circuits have been realized on banknotes, for anti-counterfeiting applications[27]. It was shown that despite the surface roughness of banknote paper, OFETs operating at less than 1 V with mobilities of 0.2 cm^2/Vs could be fabricated reliably. A photo of a circuit printed on a banknote is shown in Fig. 2b. Electronic circuits on paper have been the subject of a recent extensive review[28]. Paper substrates have been used for flexible electrowetting displays[29], as well as thermochromic displays for disposable consumer products[30]. Encouraging performance has been demonstrated for paper-based organic photovoltaics. Recently, fully roll-to-roll solution printing using gravure and flexographic printing techniques was utilized to make printed-paper photovoltaics[31]. These devices use an inverted design, with a printed ZnO/Zn back electrode and a conducting polymer transparent top electrode. Such solar cells combine ultra low-cost materials, high-throughput low-temperature roll-to-roll printing, and a flexible final product. A photo and cross-sectional diagram of these paper solar cells are shown in Fig. 2c. Another reported approach for paper-based photovoltaics features thin semitransparent paper as a substrate, with a conducting polymer transparent electrode, organic active layer, and reflective back electrode all fabricated via low-temperature chemical vapor deposition[32]. Arrays of these devices are shown in Fig. 2d, with Fig. 2e illustrating folded devices. The cells can be folded repeatedly with no degradation of the array performance.

Fig. 1 Pictographic example of electronic waste. Electronic products bring pleasure and comfort to consumers, but ultimately will reach the end of their life. Plastics have a slow biodegradation route that lasts up to 1000 years, while the electronic circuits contain many toxic and environmentally dangerous materials. Image courtesy of Park Howell (http://parkhowell.com/). Reproduced with permission.

Another natural material with a long history is silk. Silk is a polypeptide polymer, consisting of two main proteins: fibroin and sericin. Fibroin is made up primarily of repeating units of glycine, serine, and alanine that afford interchain hydrogen bonding, providing the mechanical robustness of silk fibers[33]. A molecular structure of fibroin is shown in Fig. 3a. This material combines many advantages for biodegradable or biomedical

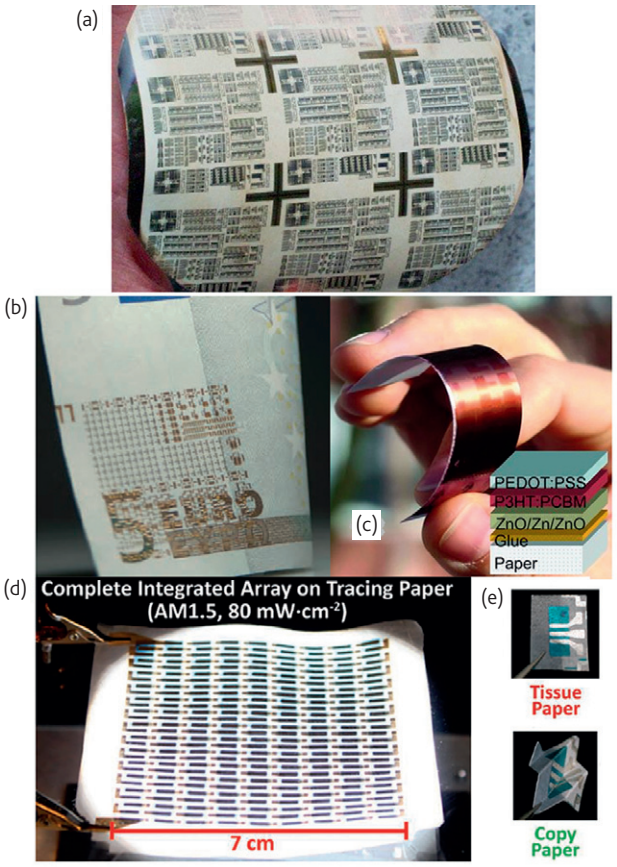

Fig. 2 Organic electronic devices fabricated on paper substrates. (a) OFET array deposited on a paper substrate. Reprinted with permission from[25]. Color image courtesy of Hagen Klauk. Copyright 2004, American Institute of Physics. (b) OFETs printed on a banknote. Reproduced with permission from[27]. Copyright Wiley-VCH Verlag GmbH & Co. KGaA. (c) A fully solution-processed roll-to-roll printed solar cell on paper. Reproduced with permission from[31]. Copyright Wiley-VCH Verlag GmbH & Co. KGaA. (d) Solar cells fabricated using a monolithic integrated chemical vapor deposition (CVD) process onto semitransparent paper, (e) examples of CVD-prepared solar cells. Repeated folding of devices did not result in degradation. Reproduced with permission from[32]. Copyright Wiley-VCH Verlag GmbH & Co. KGaA.

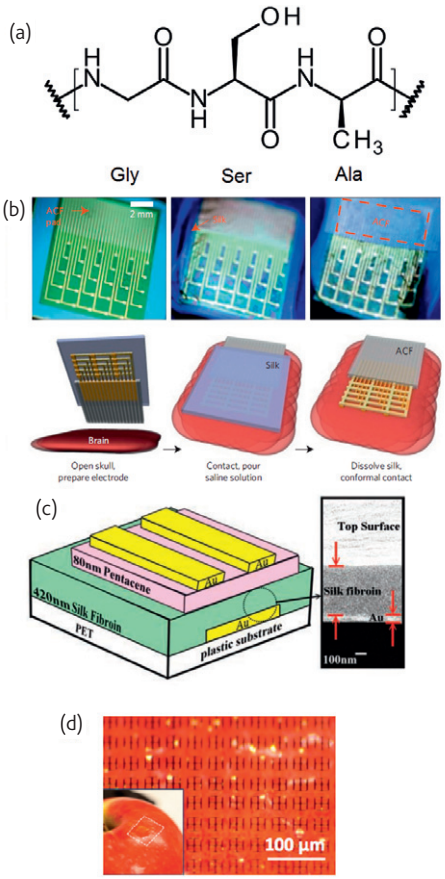

Fig. 3 Silk in electronic devices. (a) Chemical structure of silk fibroin, a polypeptide. (b) Example of a sensor array for neural recording fabricated on a silk substrate. Once the device is applied onto the brain, silk is safely dissolved and resorbed, resulting in a conformal coating of the sensor array onto the brain tissue. Reprinted from[34] by permission from Macmillan Publishers Ltd, copyright 2010. (c) Solution-processed silk fibroin as a gate dielectric for a flexible OFET. This device was reported with a mobility of ~23 cm^2/Vs. Reproduced with permission from[36]. Copyright Wiley-VCH Verlag GmbH & Co. KGaA. (d) A silk-based passive rf-ID tag, applied on an apple skin to function as a food quality monitor. Reproduced with permission from[37]. Copyright Wiley-VCH Verlag GmbH & Co. KGaA.

applications. Silk is fully bioresorbable and elicits no immune response, and thus can be safely implanted into the body. A recent study showed that an ultrathin electronic sensor array can be fabricated on silk, which can then be placed *in vivo* onto exposed brain tissue. The silk safely dissolves and resorbs, resulting in conformal coating of folded brain tissue with the sensor array (Fig. 3b)[34]. Silicon-based electronics can also be fabricated onto silk, and the silk can be used as a bioresorbable carrier to introduce the electronic element *in vivo*[35]. Silk can function as an effective solution-processed gate insulator for OFETs (Fig. 3c), supporting very high mobilities of ~23 cm^2/Vs in pentacene combined with low-voltage operation[36]. Recently, silk has been used as a substrate for passive rf-ID circuits that can be integrated directly onto food, i.e., apples, eggs, etc., as sensors of food quality (Fig. 3d)[37]. Additionally, silk is fully biodegradable and can be engineered to degrade under desired conditions, enabling targeted drug storage and delivery, e.g,[38-40].

Another protein-based material is gelatin, used commonly for capsules for oral drug ingestion. It is also fully biocompatible and biodegradable. Electronics built on hard gelatin may easily be ingested for specific biomedical applications targeting short interrogation time. Fabrication of OFET devices directly onto hard gelatin capsules has been demonstrated[41]. The protein albumin, from chicken egg whites, has been shown as a high performance cross-linkable solution processed material for OFET dielectric[42]. Protein-based materials for sustainable applications have been recently reviewed[43].

Aside from protein-based polymers, polysaccharides can also be used as biocompatible substrate materials. Polymers made from starches and polylactic acid have recently been commercially mass-produced as biodegradable plastics[6]. An example is Ecoflex (BASF), a foil plastic produced from potato and corn starch and polylactic acid. Ecoflex degrades in compost in six months without leaving any residue[44]. Caramelized glucose was recently explored as an exotic substrate for electronics; despite its sensitivity to moisture, the film forming characteristics of

ISSN:1369 7021

glucose rivaled those of glass in terms of smoothness[45]. Another 'historic' natural polymeric material is shellac. This resin is naturally produced by female lac beetles, and is harvested from trees in India and Thailand. Chemically, it is a natural polyester copolymer of terpenic and aleuritic acids, and can be processed from various polar organic solutions, such as ethanol. Advantageously, shellac can also be synthetically fabricated in a multitude of compositional grades and shades[46,47]. Both silk and shellac have excellent surface smoothness (rms < 1 nm, rivaling glass) when deposited as thin films. Shellac can easily be cast to produce substrate foils (200 – 500 μm thickness), which have been used to make OFETs and complementary-type circuits with the natural semiconductor indigo[48].

Deoxyribonucleic acid, or DNA, is the building block of life on earth. DNA is extracted in large amounts from waste products of the fishing industry, and is thus a 'natural feedstock' material that can be produced on an industrial scale. This fascinating molecule has inspired many researchers to apply it in practical applications in photonics and organic electronics. DNA can be processed from water solutions to produce films with excellent optical transparency from 400 nm through the NIR region. Recently, organic light emitting diodes (OLEDs), nonlinear optoelectronic modulators, and photonic arrays based on DNA have been reported[49-54]. DNA can also be applied as a gate insulator in organic field effect transistors. Solution-processed and cross-linked DNA was successfully implemented as a gate dielectric layer for low operating voltage OFETs[55-57]. The individual nucleobases (guanine, adenine, thymine, and cytosine) are extracted commercially for medical and cosmetic applications and have been implemented as gate dielectrics for OFETs[45]. An example of a 'green OFET' is shown in Fig. 4a. In this device the substrate is caramelized-glucose, guanine, and adenine form the gate dielectric, and a nontoxic textile dye, indanthrene yellow G is the organic semiconductor. A similar device is shown in Fig. 4b, where guanine and adenine are used as the gate dielectric for an OFET fabricated on a gelatin capsule. A thin film of adenine used in combination with electrochemically-grown aluminum oxide dielectric and C_{60} fullerene can afford OFETs with a low operating voltage (~0.5 V) and high semiconductor mobility (~5.5 cm^2/Vs)[45,58]. This summary of natural substrates and dielectrics shows that nature offers an affluence of materials choice that could be integrated into various organic electronic devices, offering alternatives for biocompatible, biodegradable, and even bioimplantable and bioresorbable applications.

Semiconductors

Nature is replete in π-conjugated molecules that can be used as semiconductors. Additionally, the synthetic dye industry produces many conjugated organic dyes that have been determined safe and nontoxic for use as food or textile colorants and inks. Compared to the exploration of biocompatible materials such as insulators and substrates, investigations into biocompatible semiconductors remain sparse. The carotenoids, such as β-carotene, are linear π-conjugated molecules that act as hole-transporting semiconductors. Devices using β-carotene and natural gate insulators such as glucose fabricated on biodegradable plastic substrates are demonstrations of truly 'natural' OFETs[41,45]. Nevertheless with mobilities in the range of 1×10^{-4} cm^2/Vs those devices are not efficient. Attempts to utilize solution-processed β-carotene in solar cells showed only modest performance[59]. The first reports of fully 'green' OFET devices

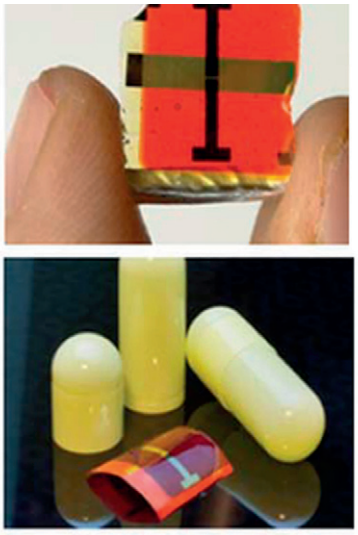

Fig. 4 OFETs on natural substrates. (a) An entirely 'bio-OFET' utilizing a caramelized-glucose substrate, Al gate electrode with an adenine/guanine gate dielectric, and the nontoxic textile dye indanthrene yellow G functioning as the semiconductor. (b) An OFET fabricated on a gelatin capsule, utilizing adenine/guanine gate dielectric and a cosmetic perylene bisimide dye. Reproduced with permission from[41]. Copyright Wiley-VCH Verlag GmbH & Co. KGaA.

featured biocompatible substrates and natural dielectrics and employed nontoxic synthetic textile dyes such as anthraquinones and perylene bisimides; these devices demonstrated mobilities in the 10^{-2} – 10^{-1} range[41,45]. Fully biomaterial-based OFETs with ambipolar charge transport mobilities in the range 10^{-2} – 0.4 were demonstrated with indigo and its derivatives[48,59-61]. In such devices, as shown in Fig. 5, substrate, dielectric, and semiconductor are all of natural-origin.

Indigo is the most mass-produced dyestuff worldwide, primarily used for the coloring of blue jeans. Though today produced synthetically, it originated from several species of plants and has been extracted and used as a dye since ancient times. Indigo, and its brominated derivative, 6,6'-dibromoindigo were arguably the subject of the world's oldest chemical industry, being produced from natural sources and prized as commodities as valuable as gold[62-64]. Their structures are shown in Fig. 6. Several indigo derivatives are present in nature, in both plants and animals, and indigo itself has been reported to be biodegradable and non-toxic[65] while also having a biosynthetic route involving strains of bacteria[66,67].

Indigo and its derivatives are thermally- and photochemically-stable molecules due to intra- and intermolecular hydrogen bonding between amine hydrogens and carbonyl groups. The excellent planarity of the molecule and H-bonding result in tight π-stacking between neighbors, with an interplanar spacing of ~3.4 Å[62]. Indigo 'breaks the rules' of traditional molecular organic semiconductors as it has minimal intramolecular conjugation, with carbonyl and amine groups seen to interrupt conjugation in the resonance model. However, the excellent charge transport properties of indigoid dyes are attributed to the strong intermolecular interactions of π-stacking reinforced by hydrogen bonding. Due to the directionality of π-stacking (typically along the crystallographic *b*-axis for most indigoids) charge transport is highly anisotropic. In order to achieve good OFET performance, molecules

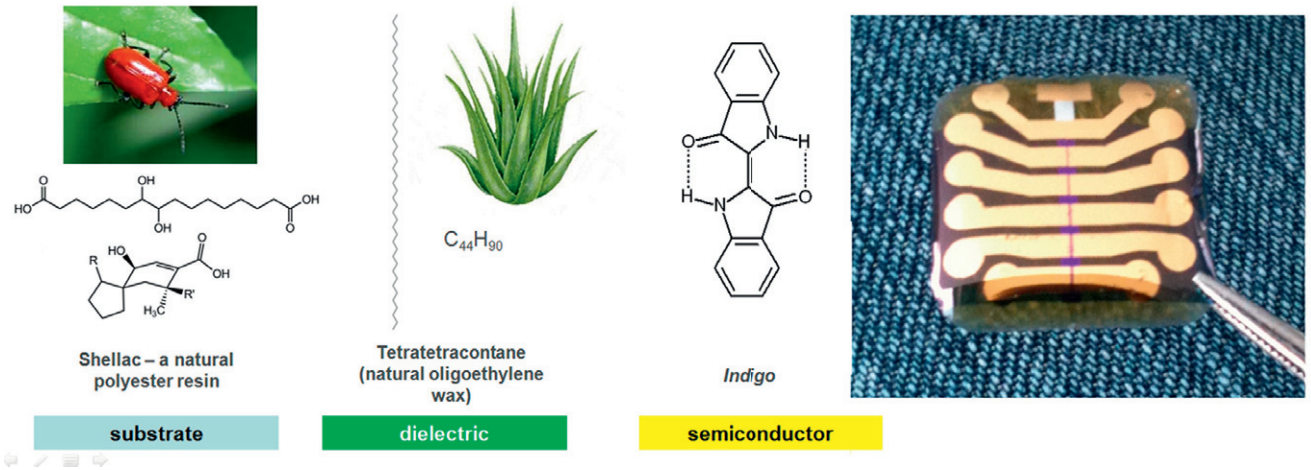

Fig. 5 An OFET device utilizing all natural materials. The substrate is shellac foil, produced by drop casting from ethanol solution. An electrochemically-grown Al_2O_3 layer passivated with the natural oligoethylene tetratetracontane serves as the gate dielectric. The semiconductor material is indigo. The image on the right shows a complete array of several such OFETs on a shellac substrate. These devices showed excellent stability and balanced electron and hole mobilities of 0.01 cm²/Vs. From E. D. Glowacki, M. Irimia-Vladu, "Natural and Nature-inspired Materials in Organic Electronics", SPIE Newsroom 2012, doi: 10.1117/2.1201201.004054. Reproduced with permission.

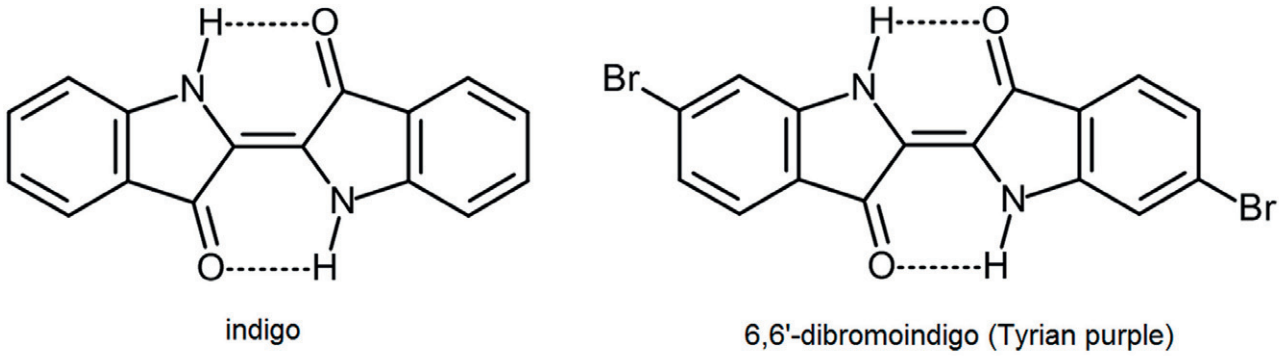

Fig. 6 Chemical structures of indigo and its dibromo derivative. Indigo is derived from several species of plants, Tyrian purple originates from the glands of different species of marine mollusks.

must adopt a 'standing-up' conformation, with π-stacking parallel to the gate dielectric. To achieve this, aliphatic dielectric materials are used, such as polyethylene or the natural oligoethylene tetratetracontane. Indigo and Tyrian purple both show reversible two-electron reduction and oxidation electrochemistry, and have small band gaps (1.7 – 1.8 eV) and thus are suitable for ambipolar OFETs and voltage inverter circuits[48,60]. An example of high performance ambipolar transport in OFETs is displayed in Fig. 7a where Tyrian purple (6,6'-dibromoindigo) is evaporated on a polyethylene-passivated aluminium oxide dielectric. The device shows well-balanced electron and hole transport channels with mobilities of 0.3 – 0.4 cm²/Vs[61]. Fig. 7b shows complementary like inverters fabricated with a Tyrian purple channel and Au source and drain electrodes. The gain of ~250 – 290 is among the best reported for a single semiconductor with a single type of contact electrode[60]. These results show that cheap and nontoxic materials of natural origin can compete with the best synthetic organic semiconductors. Research into the biodegradation and biocompatibility of organic semiconductors remains very limited.

Conductors

Exploration of biocompatible conducting materials in recent years has been a vibrant field. In addition to electronic conduction, many materials with a bio-origin are ionic conductors. Both modes of conduction have potential application in biodegradable electronic products as well as biomedical devices. Historically, the earliest organic electronic 'device'[68], a resistive switching element, was based on melanin, a biological polymeric material responsible for brown-black pigmentations in animals, including humans. Since the first reports on conductivity in melanin, it has been employed in various sandwich diode type devices[69,70]. The conductivity of melanin is heavily dependent on hydration of the material. Recently it was found that it can be used in thin-film form as a conductor in biomedical applications, showing both compatibility with living tissue and bioresorbability[71]. Originally, an amorphous semiconductor model was applied to understand the mechanism of charge transport in melanin, however more recently concise evidence has been shown that in fact proton conductivity is the mechanism responsible for charge transport in melanin[72]. Proton-conducting materials, extensively researched for

ISSN:1369 7021

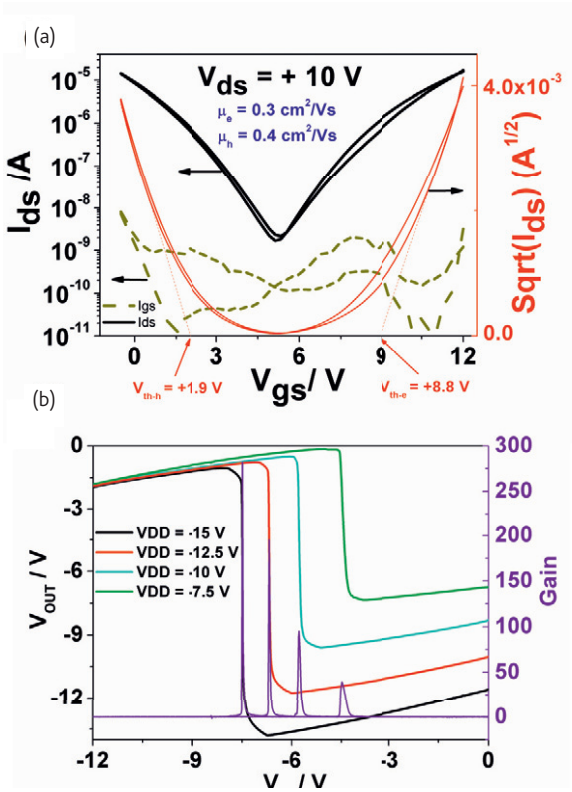

Fig. 7 Tyrian purple electronic devices. (a) Tyrian purple OFET transfer characteristics. Reproduced from [61] Copyright 2010, with permission from Elsevier. (b) Output characteristics of a Tyrian purple-based voltage inverter (a complementary-like circuit) showing among the highest-reported gains for a single organic ambipolar material. Adapted from[60]; used in accordance with the Creative Commons Attribution 3.0 Unported License.

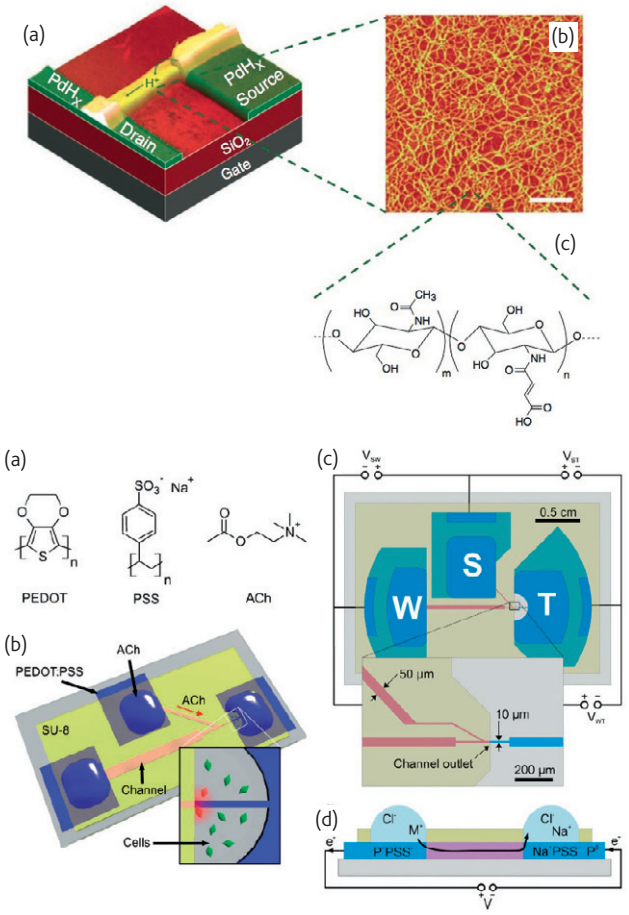

Fig. 8 (Top) A schematic of a bioprotonic transistor. Palladium functions as a protode (proton-source and drain material) with an electronic gate modulating the protonic current through chitosan, a biopolymer. Proton mobility was found to be ~4 × 10⁻³ cm²/Vs. Reprinted from[73] by permission from Macmillan Publishers Ltd, copyright 2011. (Bottom) An organic electronic ion pump, fabricated using conventional microfabrication techniques. A single device has dimensions of 10 µm. Reproduced with permission from[80]. Copyright Wiley-VCH Verlag GmbH & Co. KGaA.

fuel cell applications, have recently been recognized for great potential in biocompatible electronics. The motivation is two-fold: firstly, natural-origin proton conducting materials enable sustainable devices, and secondly, a multitude of biological pathways involve protons, therefore proton/electronic interfaces are of interest for creating biomedical devices. Many conducting polymers are uniquely suited as bioelectronics interface materials because they can conduct both ionic and electronic currents. This is not accessible for traditional metallic conductors. A recent demonstration of a proton-conducting polysaccharide thin-film transistor device controlled by the electronic field effect of a gate is a functional realization of the electronic/protonic interface[73]. A device schematic is shown in the top portion of Fig. 8. This device utilizes the polymer chitosan, obtained from the deacetylation of chitin, the structural polymer composing the exoskeletons of crustaceans. Commercially available chitosan is derived from shrimp. A recent report has shown that transistors with solution-processed chitosan proton conductors could be fabricated on paper substrates[74].

While applied research with truly 'natural' conductors remains limited, the field of synthetic conducting polymers is relatively mature. Conducting polymers such as polyaniline, poly(pyrrole), and poly(thiophenes) have demonstrate excellent biocompatibility in biological applications[15-18,22,75]. Of the well-known electron-conducting polymers in organic electronics the system poly(3,4-ethylenedioxythiophene) doped with the polyanion poly(styrenesulfonate) (PEDOT:PSS) has been implemented in a variety of biosensing applications, and even *in vivo* studies. Reports on cell growth on PEDOT:PSS films suggest a lack of any toxicity. PEDOT:PSS can be applied on living brain tissue as conformal polymer electrodes for *in vivo* electrocorticography, showing a superior signal/noise ratio compared with traditional measurements[20]. Interfacial conducting PEDOT nanotubes have also been successfully used for neural recording[76]. Recent work shows that PEDOT can even be electro polymerized *in situ* in a living brain, accomplishing a therapeutic effect[77]. PEDOT has been shown to be an effective conductor of anions as well, while PSS can serve as a conducting medium for cations such as Ca^{2+}, Na^{+}, K^{+},[78,79] and the neurotransmitter acetylcholine[80,81]. A PEDOT-based acetylcholine voltage-driven 'pump' is shown in the bottom portion of Fig. 8. A sizeable body of work has been published where PEDOT:PSS formulations function as a ionic/electronic interface material to transduce ionic currents into electronic ones and vice versa, and has been recently reviewed[15,22]. A number of other conducting polymers have been found to be biocompatible, including polyaniline and

polypyrrole[82]. Both retain conductivity and favorable mechanical properties like flexibility in biological systems while being nontoxic and not triggering an immune response. Though these common conducting polymers have been shown to be nontoxic and remarkably biocompatible, there is a lack of reports in the literature concerning biodegradation of these materials.

Conclusions

Organic materials are uniquely suited to produce electronics that can not only be sustainable and biodegradable, but can also have functionalities inaccessible to standard crystalline semiconductors, such as the functionalities required in many biomedical applications. Research in the field of the biointegration of electronics is proceeding swiftly, primarily because organic materials offer unique advantages. The consideration of biodegradability and sustainability of organic-electronic based consumer devices is still in its infancy at present. Recent demonstrations of high-performance organic electronics based on biomaterials have shown that truly 'green' electronics have potential and, hopefully, are poised to make a positive impact in the future. mt

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ISSN:1369 7021