

From Single Molecules to Thin Film Electronics, Nanofibers, e-Textiles and Power Cables: Bridging Length Scales with Organic Semiconductors

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Organic semiconductors are the centerpiece of several vibrant research fields from single-molecule to organic electronics, and they are finding increasing use in bioelectronics and even classical polymer technology. The versatile chemistry and broad range of electronic functionalities of conjugated materials enable the bridging of length scales 15 orders of magnitude apart, ranging from a single nanometer (10^{-9} m) to the size of continents (10^6 m). This work provides a taste of the diverse applications that can be realized with organic semiconductors. The reader will embark on a journey from single molecular junctions to thin film organic electronics, supramolecular assemblies, biomaterials such as amyloid fibrils and nanofibrillated cellulose, conducting fibers and yarns for e-textiles, and finally to power cables that shuffle power across thousands of kilometers.

that sound recycling solutions are needed, which become more important with increasing length scale because of the volume of material involved.

We will start our journey with the field of single molecule electronics, which promises to preserve Moore's law by enabling electronic devices the size of individual molecules. Thin film organic electronics, instead, uses active layers composed of many molecules to realize traditional devices ranging from transistors to light emitting diodes and solar cells. The active layer of thin film devices is only on the order of 100 nm, but their length can reach 100 m or more in case of roll-to-roll manufac-

1. Introduction

Organic semiconductors are touted as a unique class of materials for a multitude of emerging applications ranging from new ways to harness, store, and transport energy to new platforms for electronics based on single molecules, self-assembling biomacromolecules, or textiles. Relevant length scales range from the size of a single molecule to the length of polymer fibers and ultimately the range of power transmission lines, which can span whole continents. In this article we intend to provide a flavor for the many seemingly disparate architectures that can be realized with the help of organic semiconductors, allowing to bridge length scales from 1 nm to the size of our planet earth (Figure 1). To fully harness this immense potential of organic electronic materials, we argue

tured solar cells. We then discuss how self-assembly of many molecules may one day allow the in-situ growth of complex machinery and electronic circuits, driven by electrochemistry. Biomacromolecules including DNA, proteins and cellulose are able to form a wide range of hierarchical structures that, when decorated with conjugated polyelectrolytes, are able to conduct electronic as well as ionic charges across the length scale of nanometers to micrometers. At the same time, strong dispersion forces in the form of π -stacking as well as electrostatic interactions allow a wide range of conjugated materials to self-assemble into well-defined nanofibers that can be used to bridge contacts a few micrometers apart, hence forming, e.g., the channel of a field-effect transistor. One platform with exceptional allure is textile materials that, once equipped with electronic functionality would allow to blur the interface between user and electronics. Electrically conducting fibers and yarns prepared with conducting plastics can be woven or knitted into a plethora of complex 2D and 3D architectures that can function as keyboards, sensors, logic circuits, and displays the size of millimeters to meters. Eventually, we will discuss how organic semiconductors may become a powerful new type of plastic additives that vastly improve our means to shuffle energy and information across the distance of whole countries and continents. Finally, we postulate that first attempts to ready technologies such as organic solar cells for space may one day allow us to venture beyond our planet and explore distances—with the help of organic semiconductors—that are today safely within the realm of science fiction.

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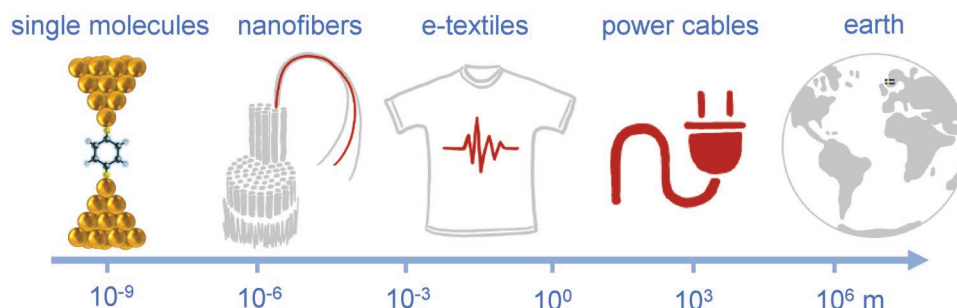


Figure 1. Organic semiconductors can (help to) bridge length scales ranging from the size of single molecules (nm) to the length of nanofibers (μm), yarns for e-textiles (mm to m), power cables (km), and finally our planet earth.

2. Single Molecule Electronics

We begin our journey of length scales by discussing the smallest possible constituents: single organic molecules. The idea of using single molecules as building blocks for electronic circuitry dates back more than 50 years.^[1,2] Originally the vision was to develop functional electronic systems, but a rapid development of silicon-based technologies has meant that, with a few exceptions,^[3] molecular electronics has remained an interesting research area mainly for the academic community. One of the challenges with single organic molecules is their minuscule size of typically 1–3 nm in length, much shorter than what can be addressed using classical top down approaches. During recent years, several test beds for the investigation of transport through single molecules have evolved such as mechanical break junctions,^[4] scanning tunneling break junctions,^[5] and others.^[6,7] Recent developments enable more parallelized fabrication of nanoscale junctions.^[8–11] These test beds have enabled scientists to develop and explore molecules acting as wires,^[12] diodes,^[13] rectifiers^[14] and switches.^[15] Initially, it was relevant to study whether molecular wires in fact were conducting.^[12] Today both systems with very high conductivity^[16] and extremely insulating ones^[17] have been identified, and the use of molecular switches has resulted in the development of compounds capable of modulating current.^[18] On the molecular scale, several different transport phenomena can take place. For many systems, coherent tunneling is the dominant means of transport, whereas for others, a sequential incoherent electron transfer is more important. In the context of materials design, one may ask: what does it take to modulate between the two different transport regimes? The simple answer is: a single atom. In a paper by Danilov et al., two of the shortest possible oligo-phenylene-vinylene oligomers, both end-capped by thiols, were placed in a three-electrode contact geometry.^[19] One molecule was bearing the thiol groups directly in the fourth position of the phenyl groups, whereas the other had a benzylic substitution pattern, thus breaking the π -conjugated structure with a single methylene (CH_2) spacer group. Interestingly, the fully conjugated system featured tunneling-based conductivity in the μS range whereas the transport through the molecule with CH_2 spacers featured sequential electron transport (hopping) and Coulomb blockade at almost three orders of magnitude lower conductivity (nS). This experiment illustrates that the interface geometries, down to the fine structure of single atoms may have tremendous influence on electron

transport at macroscopic length scales. In the context of length scales, an important question to ask is whether the transport mechanism is dominated by tunneling or by hopping. In other words, for conjugated oligomers, when does a system behave like a small molecule and when is it behaving like a conjugated polymer?^[20,21] Frisbie and co-workers set out to study this by performing transport studies of a series of conjugated oligo-phenyleneimine molecular wires (**Figure 2**).^[20] The authors found that for oligomers up to 4 nm long, the transport was temperature independent and thus dominated by tunneling, whereas for longer oligomers transport occurred through

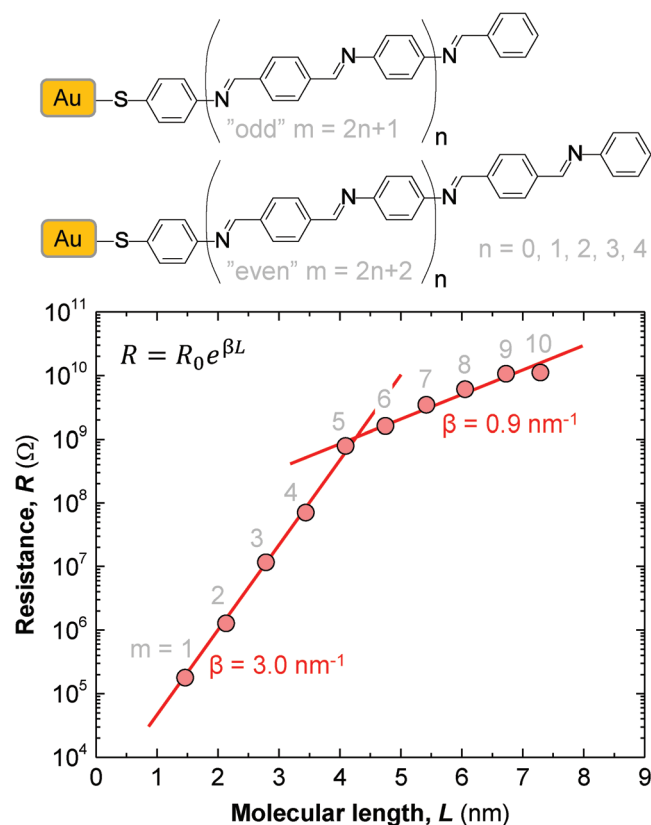


Figure 2. Resistance of oligophenyleneimine molecular wires as a function of molecular length; the transition from tunneling to hopping transport occurs around $m = 5$. Data and chemical structures adapted with permission.^[20] Copyright 2008, American Association for the Advancement of Science.

thermally assisted hopping. These observations of transport on the nanoscale highlight the importance of controlling both the interface and molecular architecture when designing materials with tailored electronic properties.

3. Thin Film Organic Electronics

In analogy to charge transport through single molecules, the constitution and conformation of organic semiconductor molecules has a profound impact on the optoelectronic properties of solid state materials that form the active layer of thin film devices such as transistors, light emitting diodes and solar cells. On the molecular level, the opto-electronic properties of conjugated polymers, for instance, are determined by three characteristic lengths: 1) the persistence length, i.e., the chain stiffness that is determined by the choice of repeat unit, 2) the side chain length, and 3) the main chain length, which is given by the degree of polymerization or molecular weight. The persistence length can range from only a few nanometers in case of polythiophenes to 20 nm in case of polyparaphenylenes and diketopyrrolopyrrole based copolymers,^[22,23] and for example impacts the degree of optical absorption and hence light harvesting efficiency of polymer solar cells.^[23] Side chains are necessary to induce both processability from solution and melt. Typically, there is an optimal length for which solubility and opto-electronic performance are balanced, e.g., hexyl side chains are preferred in case of poly(3-alkylthiophene)s.^[24] The molecular weight of poly(3-hexylthiophene) (P3HT) can reach several 100 kg mol⁻¹ (end-to-end distance 240 nm), and determines the lamellar thickness of P3HT crystallites as well as the (electrical and mechanical) connectivity of the material through tie chains and entanglements, which in turn impact the charge carrier mobility measured with field effect transistors^[25,26] and the power conversion efficiency of polymer solar cells.^[27,28] As a result the field effect mobility of P3HT strongly increases with molecular weight from 10⁻⁶ to 10⁻¹ cm² V⁻¹ s⁻¹; other (more rigid) conjugated polymers today routinely exceed 1 cm² V⁻¹ s⁻¹.

For many applications mixtures of two (or more) semiconductors are used to facilitate charge transfer, which occurs across a distance of typically less than a nanometer since the wavefunctions of the donor and acceptor must overlap. In bulk-heterojunction organic solar cells excited states are generated through the absorption of light, and undergo charge transfer at a donor:acceptor interface (followed by dissociation into free charge carriers), giving rise to an open-circuit voltage that is determined by the resulting charge transfer state.^[29] Similarly, in doped systems ground state charge transfer occurs between dopant molecules and organic semiconductors, and gives rise to conducting materials that can be used as interlayers in thin film devices, and are of interest for organic thermoelectrics.^[30] The phase behavior of bulk-heterojunction blends and dopant:semiconductor mixtures determines to which extent a miscible phase can be generated, where molecular contact permits charge transfer. Phase separated domains, whose size can range from tens of nanometers to micrometers, impede charge transfer but can benefit charge transport, especially in organic solar cells. In case of immiscible mixtures, or mixtures that tend to undergo crystallization, it is possible to kinetically trap a

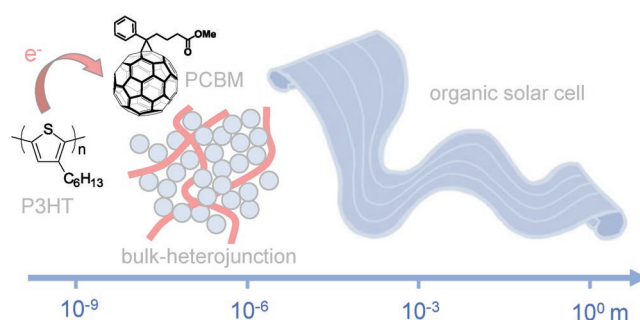


Figure 3. Length scales in an organic solar cell range from 10⁻⁹ m, here exemplified by charge transfer between P3HT and a PCBM acceptor, to the size of phase separated domains in a bulk-heterojunction blend, which are ideally on the order of not more than 10⁻⁸ m but can be as large as 10⁻⁶ m, and finally the length of a roll-to-roll manufactured photovoltaic panel, which can exceed several meters.

fine nanostructure by exploiting the typically high glass transition temperature of conjugated polymers.^[31]

The dimensions of thin film electronic devices, which typically consist of multilayer stacks that are deposited on planar (and possibly flexible) substrates, constitute a further set of length scales. In single junction organic solar cells, for instance, an active layer composed of a bulk heterojunction blend vertically bridges a length scale of typically about 100 nm, and connects to the transport layers and electrodes in a sandwich configuration. The size of a solar cell, however, can range from millimeters in case of lab scale devices, to large-area panels that are manufactured in a continuous roll-to-roll process,^[32,33] and whose length (of up to at least 100 m) is only limited by the dimensions of the flexible substrate foil (**Figure 3**). Manufacturing of organic solar cells is predicted to be particularly cost-effective, resulting in a payback time as low as one day,^[34] which—as pointed out by Inganäs and Admassie^[35]—makes them an intriguing contender for the electrification of continents such as Africa that currently lack electrical grids, where decentralized grid designs are a viable alternative. Power generation with (organic) solar cells could be complemented with storage solutions based on wood batteries^[36] or molecular solar thermal energy storage systems.^[37]

Since *plastic* electronics are envisaged to be used in large volumes for a myriad of applications, ranging from large area solar cell installations to radio frequency identification (RFID) tags that form the backbone of the *Internet of Things*, recycling is a major concern. Ubiquitous use will only become a reality if solutions can be found that do not add to the growing mountain of plastic waste, but instead target some of the *UN Sustainable Development Goals*, for instance by enabling a more resource efficient use of materials, food, energy, water etc.

4. Organic Nanofiber Electronics: A Biotemplating Approach

One intriguing hierarchy in terms of size are fiber-like electronic structures such as filaments with diameters ranging from several nanometers to tens of nanometers, with a length spanning up to tens of microns. These nanofibers are of

fundamental interest^[38] for three reasons: 1) their length provides a means to bridge the realm of organic nanoelectronics and micrometer-sized conventional electronics,^[39] 2) their high aspect ratio allows folding of single fibers or fiber bundles into complex 3D nanoshapes,^[40,41] and 3) their dimensions allow them to be dissolved or dispersed in liquid media, preferably water, which enables higher order supramolecular self-assembly.^[42]

Although a rich library of organic semiconductors and conjugated polymers is available and, quite a few processing routes and materials systems have been developed,^[38,43–49] our ability to precisely program these materials into processable, ordered nano-to-micrometer 1D structures is still very limited. A recent example includes the self-assembly of diblock copolymers into submicrometer thick nanofibers with a mobility in the range of $10^{-3} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ ^[50] which, however successful, illustrates the relatively complex synthetic chemistry that is required to attain the programmed capability for self-assembly.

A viable alternative for supramolecular self-assembly instead relies on nature, which has already produced endless possibilities for programmed assembly of biomolecules and biological structures. Conjugated polymers with functional chemical groups enable simple routes for co-assembly with biological structures under aqueous conditions, to form conjugated polymer/biomolecular hybrids. These hybrids constitute a powerful platform for building electronic nanodevices, a paradigm pioneered by Inganäs and others.^[51–53] Here, we highlight examples on how the major classes of biomolecules: DNA, proteins, cellulose and lignin have been explored in self-assembled organic nanoelectronics.

DNA forms the famous double helix with a diameter of about 2 nm (B-DNA). A single double strand can contain billions of base pairs, amounting to a total length of over 1 m and an aspect ratio exceeding 10^8 . The molecule can be stretched and aligned, thus readily expanding to the micrometer scale.^[54] Some early works of decorating DNA with conjugated polymers include assembling the polymer with single stranded DNA. The hybridization of single stranded DNA with its complementary chain induces a conformational change of the polymer, therefore changing its electrochemical or optical properties.^[55–58] One particularly interesting property of DNA is its high aspect ratio at such a small length scale, which is difficult to attain through standard nanofabrication.^[59–62] The DNA template allows the fabrication of ultrathin (in the nanometer range) channels of electrochemical transistors.^[63] Single stranded DNA can also be labeled onto a conjugated fluorophore core. Upon hybridization, the triblock assembles into micrometer-sized supramolecular wires.^[64] Templating electronic materials with single long chains of DNA thus enables anisotropic properties at the molecular level. This route can for example bridge the dimensional gap between the nano- and microscopic world, enabling large-scale connection to microelectronic devices.

The combination of DNA technology and organic semiconductors is still in its infancy. Opportunities are rich and open. In the past decade, one of the most exciting developments of DNA-based technology is the emergence of *DNA origami*, where predesigned DNA sequences self-assemble into specific nanoarchitectures.^[65] This technique provides unprecedented

precision of programming matter at the nanoscale into almost any 3D shape, which can then be used as a template for assembling nanoparticles^[66] and polymers. In the near future, it is plausible that we can engineer self-assembled 3D nanoelectronics, such as antennas and metamaterials, using DNA origami as templates. A remaining challenge is the design of water-soluble conducting polymers, with metallic or semi-conducting properties. The polymers can be empowered with functional groups that interact specifically with DNA, either during or after the base-pairing process that forms specific nanostructures. This approach has recently been shown for synthetic nonconducting polymers^[67] but is yet unexplored for conjugated polymers. Alternatively, by rationally creating polymer-DNA (or oligonucleotide sequences) conjugates, the DNA origami technique can guide the assembly of conducting polymers into desired shapes.^[68] The controlled assembly of multiple conjugated polymers with different chemical structures at the molecular level is also potentially feasible with this technique.

Nature has the extraordinary ability to precisely assemble the protein units it produces into programmed, nonequilibrium complex shapes. Self-assembled supramolecular protein structures, in the shape of helices, hollow tubes, filaments, and nanoribbons, are abundant in nature. These 1D shapes typically are several nanometers in diameter and can extend to several micrometers, giving an aspect ratio of over 1000. Some of the important protein filaments include cytoskeletal filamentous actin (F-actin, helical filaments with a diameter of $\approx 8 \text{ nm}$), microtubules (hollow tubes with an outer diameter of $\approx 25 \text{ nm}$) and amyloid fibers (hollow tubes or nanoribbons, with a width of 5–15 nm).^[69–71] On the other hand, the fast development of synthetic peptide and recombinant protein engineering enables precise control over the protein sequences and chemistry, adding possibilities for carefully designing the functions and structures of the self-assembled protein supramolecular architectures.^[72–75]

Self-assembled supramolecular protein structures have provided powerful tools for guided assembly of materials.^[76] The formation of amyloid fibers can for example guide the assembly of conjugated oligomers into electroactive luminescent nanowires.^[77] Naturally derived proteins can carry hydrophobic organic dyes in their hydrophobic domains and disperse them in water. These dye-containing proteins assemble into fibers or films in aqueous medium, a route to fabricate polarized light-emitting devices.^[78] As the amyloid fibrils are closely related to several diseases, conjugated polymers that are responsive to the formation of amyloid fibrils can also be optical probes for diagnostics^[79] in vivo.

Protein filaments have diameters in the range of tens of nanometers and display superior mechanical strength. The hollow tubes, microtubules, with their relatively stiff structure, have a persistence length of over one millimeter. With the length reaching the micrometer range, metallic polymer decorated protein fibrils can be another platform for the connection to microelectronics,^[80,81] or for the fabrication of 1D devices such as nanowire transistors.^[82] Microtubules are involved in multiple biological processes. Their hollow nanotube structure represents another type of quasi-1D devices that are highly interesting yet difficult to fabricate.^[83] Decorating these protein

structures with conducting polymers can also provide means of interfacing microelectronics with living cells.^[84,85] The tubule structures are especially interesting, as they can be potentially used as tools to understand and control ion and mass transport into the cell.^[86,87] These conducting polymer decorated protein filaments also have the potential to be integrated into living cells and act as electrodes to record and modulate the cell activities.^[88,89]

Cellulose and lignin constitute the main structural components in plants. Cellulose is the most common polymer on earth with an annual biomass production of about 10^{12} tons.^[90] Some cellulosic parts of plants form complex micrometer-sized shapes such as helices, recently extracted from the xylems of celery.^[91] Once decorated with PEDOT-S (see **Figure 4** for chemical structure), a self-doped and water soluble derivative of poly(3,4-ethylenedioxythiophene) (PEDOT), these conducting helices could function as terahertz absorbers thanks to their specific size.^[91] The main form of cellulose material in the industry is wood pulp short microfibrils, which in turn are composed of bundled nanofibers, each with diameters ranging from 5–70 nm and lengths from 100 nm up to several micrometers, called cellulose nanofibrils (CNF).^[92] CNFs thus constitute our most abundant source of advanced green nanomaterials for the future. They have remarkable mechanical strength (a single nanofiber can have a strength of 3 GPa)^[93] and can be processed from dispersions into various types of nanocomposites.^[90,94] Interestingly, they also display specific interactions with pi-conjugated molecules such as carbon nanotubes^[95] and conjugated polymers such as PEDOT.^[96] As a scaffold, CNF provides a large surface area and mechanical support to the conjugated polymers,^[97] allowing them to be dispersed and to be built into high-performance hybrid devices such as supercapacitors.^[98–101] To date, however, only few studies explore the properties imparted by the high degree of anisotropy of nanocellulose, or use

individual CNF as a template^[102,103] for building conducting polymer structures. We argue that the design of conjugated polymers and CNFs for specific types of interaction can enable ultrastrong, high-aspect-ratio, electroactive nanofibrils in bulk quantities, providing a means for the self-assembly of nanoelectronics, possibly in vivo as demonstrated by Stavriniadou et al. who infused the xylems and leaves of roses with PEDOT-S and a conducting ink of CNF and poly(3,4-ethylenedioxythiophene); poly(styrene sulfonate) (PEDOT:PSS; see **Figure 4**), respectively, to create electronic plants.^[104]

Lignin is the second most abundant biopolymer next to cellulose. It is a branched polyphenolic polyether with a complex, aromatic-rich structure that varies depending on the plant species and the isolation process. Lignin has long been regarded as a by-product or waste product of the paper pulp process. The rich quinone groups in lignin are ideal redox centers for electrochemical energy storage^[105,106] in flow batteries or aqueous rechargeable batteries. The electronic insulating nature of lignin, however, bars the effective access to these redox active quinones in the electrodes. Inganäs and colleagues were pioneers in showing that conducting polymers serve as molecular wires to grant electrochemical access to the quinones in conjugated polymer:lignin polyelectrolyte complexes, e.g. using the conducting polymers PEDOT or polypyrrole.^[36,107] This approach dramatically enhanced the capacitance of conjugated polymer-based energy storage devices (the polypyrrole:lignin electrode reached a conductivity of $\approx 1 \text{ S cm}^{-1}$ and a specific capacitance of 350 F g^{-1} in water) without addition of other synthetic and expensive electrode materials. The marriage of this abundant biopolymer with conjugated polymers for energy storage may be one of the most promising areas of application. New advances in processed lignin now enable lignin nanofibers through, for example, electrospinning or self-assembly induced by ice-segregation.^[108] Lignin nano-objects, in combination

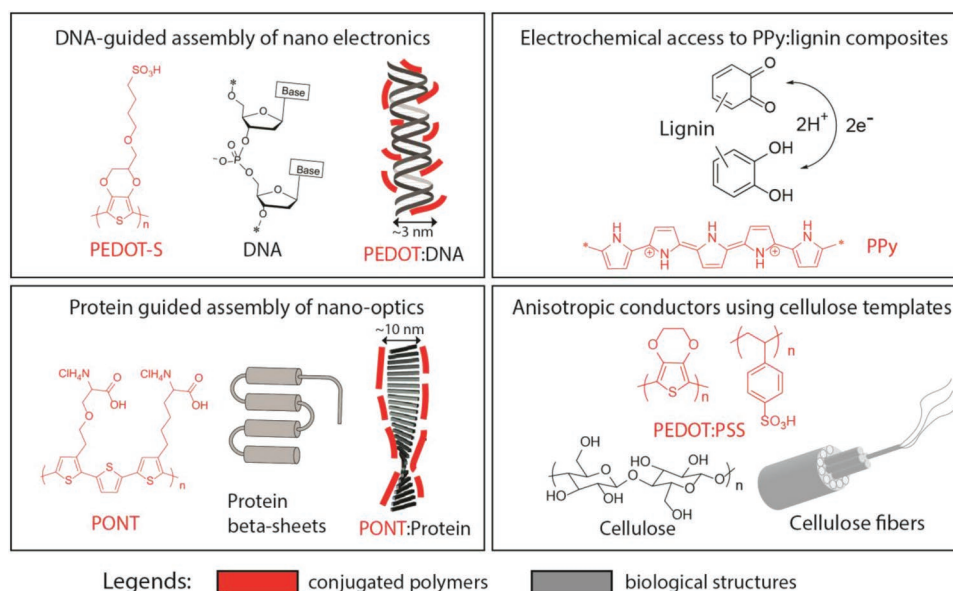


Figure 4. Conducting polymer:bionanowire hybrids: PEDOT-S assembly on DNA as conducting nanowires or ultrathin electrochemical active channels; Polypyrrole (PPy):lignin interpenetrated polymer network gives access to the redox-active quinone groups; PONT as an optical probe to detect amyloid fibril formation; and cellulose nanofibrils (CNF) stabilize and reinforce the electron and ion conducting PEDOT:PSS.

with other materials such as CNF, can enable conjugated polymer:lignin bionanocomposites with high aspect ratio, high surface area, high strength, and optimized pathways for electron and ion conduction, enabling truly low-cost and flexible energy storage systems.

5. Conducting Fibers and Yarns for e-Textiles

Textiles have found their way into every aspect of life from clothes to technical textiles used in health care, as upholstery, or for construction. The incorporation of electronic functionalities would without doubt lead to a myriad of new ways in which we can use and interact with textiles.^[109–111] Electronic textiles (e-textiles) are poised to constitute an integral part of tomorrow's *Internet of Things* consisting of innumerable miniature devices such as RFID tags. To become a versatile platform for electronics, e-textiles need to incorporate a wide range of functionalities ranging from 1) data input through, e.g., sensors and keyboards to 2) data processing using logic circuits, 3) data storage with memory devices and 4) transmission of information via antennas and displays. Such e-textile devices can then be powered by textile-based batteries and supercapacitors that are charged through energy harvesting textiles, which harness the triboelectric, piezoelectric, thermoelectric, or photovoltaic effect.^[112,113] Organic semiconductors and, more generally, conducting plastics (including even carbon black filled polyolefins) have been used to demonstrate a wide range of different devices, which are mostly built on the surface or at the junction of traditional textile fibers that then act as a passive substrate. More elaborate designs integrate (semi)conducting fibers and yarns directly into novel device architectures. Even though a wide range of individual components have been demonstrated there is a need for the development of common design standards that permit to integrate different types of devices into the same e-textile, ideally using established textile manufacturing processes.

We identify a range of length scales that need to be considered. There is the thickness of coatings, which is on the order of hundreds of nanometers to one micrometer and therefore can form the different layers of planar optoelectronic devices, e.g. the light harvesting (emitting) layer and electrodes of diodes as demonstrated for instance by Gaudiana and co-workers who realized wire-shaped organic photovoltaic solar cells.^[114] The distance between fibers can range from micrometers to millimeters, which can be bridged for example by an electrolyte that connects two conducting fibers or yarns to form an electrochemical transistor.^[115,116] Finally, there is the length of the fibers themselves, which can take the form of either a virtually endless monofilament or a multifilament yarn, and therefore bridge length scales ranging from centimeters to meters. Conducting filaments and yarns are therefore able to shuttle both power and information within an e-textile, and can act as connectors of various electronic devices.

Conducting fibers and yarns can be realized with organic semiconductors through several different approaches. Conjugated polymers such as poly(3-hexylthiophene) (P3HT) can be shaped into fibers using traditional manufacturing techniques including both solution and melt spinning,^[117,118] but

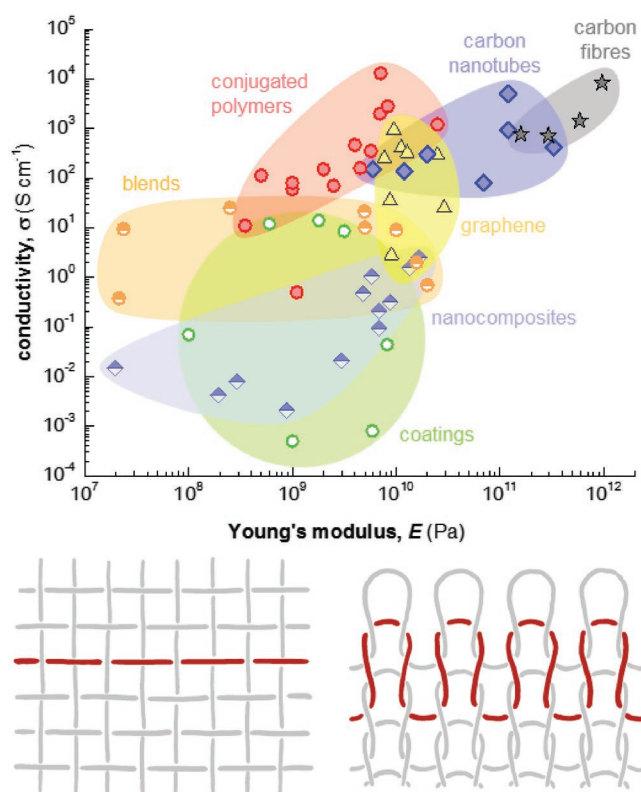


Figure 5. Top: Ashby plot of the electrical conductivity versus Young's modulus of carbon based fibers. Reproduced with permission.^[110] Copyright 2018, Elsevier. Bottom: Schematics of a plain weave and a knitted fabric.

also electrospinning, with an electrical conductivity of typically about 10^2 S cm^{-1} and in some cases more than 10^3 S cm^{-1} upon doping with, e.g., iodine or iron(III) chloride (see **Figure 5**). A binder polymer can be added to the organic semiconductor that allows to vary the mechanical response from reversibly *stretchable* in case of blends with a styrene-isobutylene-styrene triblock copolymer or polyurethane,^[119,120] to very *stiff* with a high Young's modulus of more than 10 GPa when poly(p-phenylene terephthalamide) (PPTA, i.e., Kevlar) or ultrahigh molecular-weight polyethylene is used as the matrix.^[121,122] A disadvantage is the diluting effect of the matrix material, which typically limits the electrical conductivity to about 10 S cm^{-1} (**Figure 5**), with the notable exception of wet-spun and hot-drawn PEDOT:PSS microfibers with a conductivity of more than 10^3 S cm^{-1} ^[123]—if we consider that the polyelectrolyte PSS likely also acts as a binder. Finally, it is possible to coat or dye traditional textile fibers or yarns with organic semiconductors, resulting in an architecture which readily lends itself to the fabrication of multilayer optoelectronic devices consisting of several concentric layers. Recent examples include coating of nylon monofilaments or dyeing of silk yarns using an aqueous PEDOT:PSS dispersion,^[116,124] and vapor deposition of PEDOT onto silk, linen and wool fabrics.^[125] Overall, conducting fibers and yarns with a wide range of mechanical and electrical properties can be realized using carbon based materials, chiefly conjugated polymers as well as graphene and carbon nanotubes (**Figure 5**). We note that the conductivity tends to increase

along with the Young's modulus,^[126] which arises because of the importance of alignment along the fiber axis, as this aids both charge transport and the transmission of mechanical force. Uniaxial orientation of the conjugated polymer backbone enhances the charge carrier mobility, as first reported by Inganäs and co-workers for poly(3-octylthiophene) (P3OT) that was stretch-aligned on a polyethylene support foil.^[127]

Truly functional fibers and yarns must be able to display a high degree of stability. It is essential that they can withstand the, often, harsh conditions of traditional textile manufacturing processes, where different types of mechanical stress can arise ranging from friction to flexing and tension. We argue that e-textiles will become only truly viable once established production routines can be utilized since the textile industry typically operates within tight margins. Many reports are limited to discontinuous batch fabrication schemes, instead of continuous spinning and dyeing processes, and fabric-based devices are manufactured on a lab-scale by skilled PhD students, which however is too labor intensive to be of practical relevance. Further, it is paramount that each component of an e-textile can tolerate prolonged and repeated exposure to ambient conditions, to water and to various chemical agents that are for instance found in washing powders. Further, it would be advantageous if e-textiles continue to function when wet, which we have accomplished in case of piezoelectric energy harvesting textiles where one of the electrodes is buried as the inner core of bicomponent filaments surrounded by a poly(vinylidene fluoride) (PVDF) outer sheath.^[128] We have recently reported a batch process for dyeing silk yarns with PEDOT:PSS, which resulted in a highly stable bulk electrical conductivity of 14 S cm^{-1} that was retained during at least 1000 bending cycles as well as machine washing using a commercial washing powder but also dry cleaning.^[124] Upscaling to a continuous roll-to-roll dyeing process resulted in continuous yarns with improved conductivity of 70 S cm^{-1} that we were able to process using an industrial embroidery machine or a weaving loom, to construct a keyboard consisting of a series of capacitive touch sensors.^[129]

The textile format can be used to impart additional functionality that cannot be realized with a single fiber or yarn alone. A woven textile comprises separate yarns in the weft and warp direction that form a grid where each junction can be addressed individually (Figure 5). Woven grids can then be used to define a matrix—with one electronic component at each junction—or a circuit of, e.g., resistors and electrochemical transistors that together form a logic circuit.^[116] We argue that such electrochemical transistors, fabricated by, e.g., connecting two fiber electrodes with an electrolyte (or ideally a solid polyelectrolyte), are ideally suited for the textile format. The operation voltage is less sensitive to the dimensions of the device but instead depends on the electrochemical potential of the system (typically one volt). Hence, the distance of the fiber electrodes and the shape of the electrolyte can vary without affecting the operation of the transistor, which is critical considering the relatively poor precision of textile manufacturing. An additional advantage of a woven textile is—crimp aside—the minimal bending of yarns, which facilitates the use of relatively stiff but highly conducting materials (cf. Figure 5, top). However, woven textiles are relatively rigid unless an elastic material is added. Instead, knitted textiles offer superior stretchability because

each yarn and loop can move relative to each other without deforming the yarns themselves (Figure 5). The difference in rigidity of woven and knitted fabrics allows to design textile muscles or “textuators” that either amplify force or strain.^[130]

One aspect that to our dismay receives far too little attention by the organic electronics community is the recyclability of the many new types of devices that we aim to realize. Multilayer stacks of thin films, coated on a planar substrate or on a fiber, can only be separated into their individual components with great difficulty. Heavily integrated e-textiles with their multitude of multicomponent devices and interconnects are unlikely to be compatible with the current drive toward a circular use of fabrics and clothes, making energy recovery the only viable recycling option. We propose that viable solutions should employ the least number of different components, including only a handful of organic semiconductors—prepared with environmentally friendly synthesis routes, and possibly complexed with renewable materials such as cellulose—and a minimal amount of processing and performance-enhancing additives. A recyclable e-textile could be constructed using a handful of high-mobility semiconductors that can be p- or n-doped with two suitable molecular dopants, supplemented with selected inert polymers that can act as the substrate, binder, electrolyte and encapsulation. These components could then be combined in different amounts to create semiconductors and conductors (highly doped semiconductors) with a variety of mechanical properties achieved by using different types of binders or substrates. Further, solution processability from orthogonal (and ideally green) solvents would facilitate both cost-effective production and the recovery of the individual materials once the e-textile has exceeded its lifetime. The synthesis of new organic semiconductors and the design of novel optoelectronic devices should be carried out with sustainability in mind.

6. High-Voltage Cables for Long Distance Power Transmission

Renewable sources of energy such as wind, solar and hydro power are often located far away from the end user and therefore efficient transport of electricity over long distances is vital for achieving a fossil-free economy.^[131] The ultimate goal is to be able to shuffle power across thousands of kilometers, which would enable to, e.g., use hydro power from Scandinavia to power Europe during the night, whereas during the day solar power from Southern Europe or even Northern Africa could be harnessed. This approach is particularly suitable for parts of the world that already feature well-developed electricity grids. In less developed regions that are not yet electrified, decentralized power generation with for instance (organic) solar cells represents an intriguing alternative (cf. section on thin film electronics).^[35]

The electrical conductivity of conjugated materials is too low to be relevant for the transport of power itself (cf. Figure 5, top). However, they could play an important role as additives that improve the dielectric properties of the several centimeters thick polyolefin insulation that surrounds the conducting core in high-voltage power cables (Figure 6), which would facilitate a higher transmission voltage and therefore minimize losses.

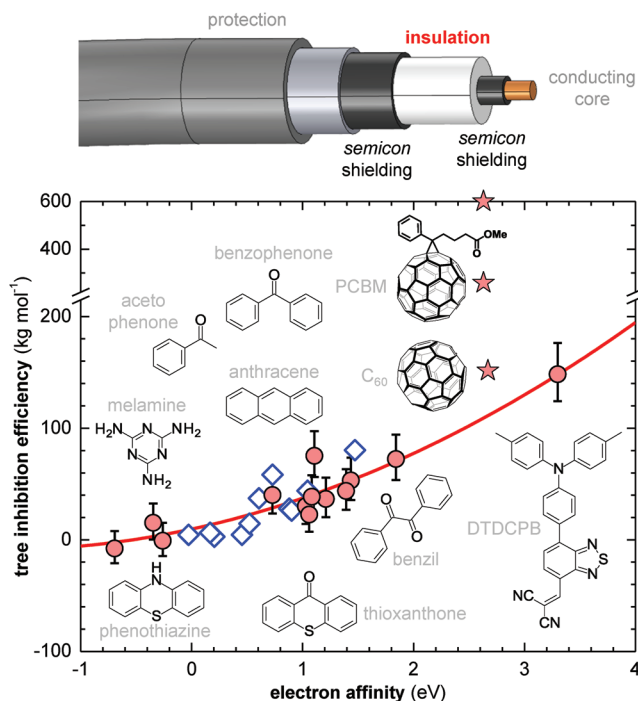


Figure 6. Schematic of a high-voltage power cable comprising a conducting core that is surrounded by semicon shielding, several centimeter thick insulation and protection layers (top); electrical tree inhibition efficiency of XLPE containing voltage stabilizers with different electron affinities, normalized with respect to the molal concentration of the additive. Adapted with permission.^[137] Copyright 2015, The Royal Society of Chemistry.

Further, organic semiconductors but also carbon nanotubes or graphene could play an important role in replacing carbon black as the conductive filler in the so-called semicon shielding layers (with a conductivity of about 10^{-4} S cm⁻¹) that sandwich the insulation layer (Figure 6).

The breakdown strength of an insulation material can be increased through the addition of so-called voltage stabilizers, i.e., conjugated molecules that scavenge high-energy electrons and as a result inhibit electrical treeing, which constitutes a major breakdown mechanism in case of high-voltage alternating current (HVAC) cables. Early studies have introduced a number of potential voltage stabilizers based on aromatic cores such as anthracene, benzophenone, benzil, thioxanthone, and even 2,4,6-trinitrotoluene (TNT).^[132–136] The correlation between the molecular weight of voltage stabilizers and their tree inhibition efficiency is counter-intuitive; a comparison of benzil type stabilizers with either methoxy, dodecyloxy, or triacontyloxy pendant chains revealed that lighter molecules are more effective, possibly because they can more easily migrate through dielectrophoresis to regions where they are most needed.^[133] We have identified a high electron affinity as a guiding criterion for the selection of efficient voltage stabilizers, and found that molecules such as DTDCPB (see Figure 6 for chemical structure)—originally developed as a donor for organic photovoltaics—are exceptionally effective in retarding the formation of electrical trees in crosslinked polyethylene (XLPE).^[137] In a further study we established that the fullerenes C₆₀ and PCBM

(see Figure 6), which are widely used as electron acceptors because of a combination of high electron affinity and mobility, are on a molal basis the most efficient voltage stabilizers that have been identified to date.^[132]

Tools that permit to further reduce the electrical conductivity of, e.g., ultraclean XLPE with 10^{-16} S cm⁻¹ are thought to be critical for the design of better high-voltage direct current (HVDC) insulation.^[138] Recently, C₆₀ has been found to increase the resistivity of polypropylene by more than one order of magnitude through the introduction of deep traps, while at the same time also leading to improved dissipation of space charges.^[139] Therefore, it can be anticipated that organic semiconductors together with the use of polyethylene blends,^[140] metal oxide nanoparticles^[141] and graphene oxide^[142] join our growing toolbox for the design of improved HVDC insulation. Ultimately, charge transport in polyethylene insulation and organic semiconductors is governed by the same physical principles^[138] albeit the concentration of ionic and electronic charge carriers is considerably different leading to a modulation in electrical conductivity by at least as many orders of magnitude as the length scales encountered in this article (note that the electronic mobility of polyethylene can be as high as 10^{-5} cm² V⁻¹ s⁻¹).^[143] HVDC power cables are uniquely suited to transport electricity across large distances of thousands of kilometers, and we predict that organic semiconductors will play their role in bridging the length scale of continents, i.e., fifteen orders of magnitude larger than the size of single molecules that we discussed at the beginning of our journey. In the (politically) distant future organic semiconductors may lead to power grids that circumnavigate earth, and possibly even allow us to travel to other planets. Organic solar cells are already being readied for space,^[144] where they may ultimately bridge length scales on the order of the size of our solar system and beyond.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

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