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Toward organic electronics with properties inspired by biological tissue

Timothy F. O'Connor, Kirtana M. Rajan, Adam D. Printz and Darren J. Lipomi*

The carbon framework common to both organic semiconductors and biological structures suggests that these two classes of materials should be easily integrated. Substantial work, however, will be required to endow synthetic electroactive materials with properties resembling those of biological tissue, which exhibits extreme elasticity, biodegradability, and the capacity for self-repair. This Highlight reviews successful integration of organic semiconductor devices with biological systems, for example, in wearable and implantable health monitors and prosthetic devices. It then points to recent work in the areas of molecularly stretchable electronics, whole devices that can degrade under physiological conditions, and conjugated polymers capable of self-healing, which together suggest the possibility of a future in which organic electronics and biological tissue can interact seamlessly.

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Tissue-like properties from organic semiconductors

The vision of organic electronics includes inexpensive,¹ flexible and stretchable² devices that can be fabricated by roll-to-roll printing³ and integrated with moving and non-planar parts of machines,⁴ buildings and windows,^{5,6} and the human body.^{7,8} Biomedical devices are regarded as an especially promising application of organic semiconductors—*i.e.*, “organic bioelectronics”^{9,10}—because of the perceived similarity of “soft” conjugated polymers and biomaterials. Despite the carbon framework common to both organic semiconductors and biological macromolecules, π -conjugated molecules have few properties in common with biological tissue.^{2,11} Conjugated polymers can be stiff (tensile modulus *ca.* 1 GPa)^{12–14} and small-molecule semiconductors have a brittleness characteristic of all van der Waals solids (crack-onset strain *ca.* 1%).¹² In contrast, biological tissue is elastic (tensile modulus \sim 10–100 kPa) and tough.¹⁵ Moreover, two additional properties of biological tissue—biodegradability¹⁶ and the capacity for self-repair¹⁷—would be desirable for biologically integrated electronics, but are not generally possessed by organic semiconductors.

The best performance so far achieved for wearable and implantable electronic devices has been—despite the inherent flexibility of organic thin films—based on composite structures of high-performance *inorganic* devices embedded in (or upon) elastomeric supports.^{18–21} This approach, exemplified by the work of Rogers and others,^{18–23} uses (as one strategy) serpentine interconnects that absorb tensile strain by local bending and twisting.¹⁸ The active components can be located at areas of

increased thickness²⁰ so that strain is channelled into the interconnects. These devices can also form the basis of “transient electronics”,²⁴ in which the active components, the interconnects, and the matrices can dissolve in programmable timescales under physiological conditions into harmless byproducts.²⁵

Despite the success of systems comprising inorganic components embedded in organic matrices, further capabilities would be accessed by a complementary approach that used solely organic components.^{26–28} That is, organics have characteristics not easily replicated in inorganic semiconductors.² These characteristics include solution-based printing,²⁹ facile tunability of optical, electronic, and mechanical properties by synthesis,³⁰ multimodal signal transduction, chemical selectivity,³¹ programmable degradation,³² and low embodied energy.² The challenges of making organic electronics softer and more responsive to chemical and mechanical stimuli are not addressed in a research strategy whose principal goal is to replicate the high semiconducting performance of inorganic materials, but are addressed in a strategy in which the advantages unique to organics are exploited to the maximum extent possible.

This paper highlights recent progress in areas of research within organic electronics that are designed to make the materials more like living tissue (interpreted broadly). The work covered may have been motivated by purposes distinct from the present goal of increasing the tissue-like behaviour of organic semiconductors (for example, making organic solar cells more resistant to mechanical failure is tantamount to making them tougher and more elastic¹¹) but the knowledge gained is independent of the motivation. We first highlight a few applications of stretchable organic electronics in electronic skin or biomedical applications, and then cover work whose goal is to increase the (1) elasticity, (2) biodegradability, and (3)

Department of NanoEngineering, University of California, San Diego, 9500 Gilman Drive, Mail Code 0448, La Jolla, CA, 92093-0448, USA. E-mail: dlipomi@ucsd.edu





Fig. 2 Molecularly stretchable organic electronics and mechanical properties of organic semiconductors. (a–c) Highly plasticized films of PEDOT:PSS can behave as piezoresistive signal transducers for detecting human motion. Reproduced with permission from ref. 51 Copyright 2015 Wiley-VCH Verlag GmbH & Co. KGaA. (d) Stretchable organic light-emitting devices maintain their luminescence at strains $\geq 130\%$. Reproduced with permission from ref. 54 Copyright 2014 American Chemical Society. (e) Biaxial stretching of P3HT produces a texture (f) in which the axis of π -stacking is oriented normal to the substrate (“face-on”). Reproduced with permission from ref. 63 Copyright 2013 American Chemical Society.

(surface wrinkling)⁶¹ to measure the tensile moduli of thin films that are difficult to assay using conventional means.¹² We have supported our measurements using a semi-empirical theory proposed by Seitz⁶² that predicts the tensile moduli through analysis of the thermal properties and the chemical structure.¹³ Among our conclusions was that the length of the alkyl pendant group (installed for increased solubility) is a key determinant of the tensile modulus.¹³ Lengthening of the aliphatic side chain decreases the number of load-bearing, main-chain carbon atoms per cross sectional area and reduces the ability of the main chains to associate. Both effects reduce the tensile modulus and the mobility of charge carriers, and thus polythiophene bearing octyl side chains is not only ten times more compliant—but also a poorer semiconductor—than polythiophene bearing hexyl side chains.²

The apparent competition between mechanical and charge-transport properties led us to synthesize block copolymers and “segmented” polymers whose blocks and segments permitted co-engineering of mechanical compliance and charge-carrier mobility.⁵⁰ We concluded that a highly elastic semiconducting polymer should exhibit three structural features: long alkyl side chains, a low percentage of fused rings in the main chain, and structural randomness (*e.g.*, branched, racemic side chains and a small degree of regioirregularity) to reduce the crystallinity.² It is not, however, sufficient that a semiconducting polymer simply accommodate strain without fracture; strain produces microstructural rearrangements in semiconducting polymers

that inevitably affect the charge-transport properties of the materials in ways that must be mitigated (or at least predicted).⁵⁸ While strain-induced alignment of chains has been known since the early days of conjugated polymers,^{64,65} mechanical deformation can also produce changes in the degree of crystallinity and texture.¹¹ For example, O'Connor and coworkers have shown an interesting reorientation in texture from edge-on to face-on in biaxially oriented films of poly(3-hexylthiophene) (P3HT, Fig. 2e and f).^{63,66} Such strain-evolved microstructure must be understood and predicted in any circumstance in which the active material undergoes deformation.

Biodegradability

Biodegradability refers to the potential of a material to be broken down in the outdoor environment or in the body without the release of toxic byproducts.^{67–69} While environmentally benign disposal of electronics is increasingly mandated by regulation, physiologically benign disintegration of implanted devices imposes even stricter constraints on the design and selection of materials. (Though a molecule whose metabolic fate is benign is unlikely to be deleterious to the environment.) Rogers and coworkers have reported electronic devices based on silicon nanomembranes that are stretchable and also soluble under physiological conditions.²⁴ Producing all-organic circuits from components that are all biodegradable is an exciting prospect,³² because the semiconductors, electrodes, and substrates could be tailored by synthesis to degrade. In an early example of a biodegradable organic electronic device, Bettinger *et al.* fabricated a thin-film transistor on a resorbable poly(lactic-co-glycolic acid) (PLGA) substrate (Fig. 3a),¹⁶ and poly(lactic acid) can also serve as a substrate for biodegradable organic devices.⁷⁰ Silk fibroin is a biologically derived material that is both highly elastic and biodegradable.⁷¹ In a recent demonstration, Liu *et al.* produced an organic solar cell (Fig. 3b–d) for which at least the substrate was biodegradable.⁷² While the fate of organic semiconductors in the body has not been widely studied, researchers may be inspired by the fact that melanin—a naturally occurring semiconducting polymer—is degraded into harmless byproducts. Melanin, however, has a low mobility of charge carriers, and despite its biological origin, is mechanically brittle.^{16,67} New synthetic systems will be required that combine favorable charge-transport properties with programmable degradation in a single material.^{73–75}

Capacity for self-repair

Self-healing behavior in non-conjugated polymers is the subject of a large and expanding literature.^{17,76} However, the vast majority of systems for self-repair of bulk polymers—*e.g.*, encapsulation of healing agents in hollow fibers and micron-sized particles—are not amenable to very thin films (≤ 100 nm) characteristic of organic optoelectronic devices.⁷⁶ We are thus interested in intrinsically healing materials that repair simply upon contact of the fractured surfaces. Materials can be self-healed by





Fig. 3 Biodegradable organic electronics. (a) Organic thin-film transistors fabricated on a biodegradable substrate decompose into byproducts that are not believed to be harmful over a period of 70 d in a solution mimicking the physiological environment. Reproduced with permission from ref. 16 Copyright 2010, Wiley-VCH Verlag, GmbH & Co. KGaA. (b) Organic solar cells fabricated on silk fibroin substrates exhibit reversible flexibility (c) and facile degradation (d). Reproduced with permission from ref. 72 Copyright 2015 American Chemical Society.

non-covalent interactions and by covalent bonds. Hydrogen bonding^{77,78} and Diels–Alder cycloadditions^{55,79} have been explored for these purposes. A prerequisite to both types of repair is a sufficient level of mobility of polymer chains to allow intermixing of polymer chains across the fractured interface.^{80,81} This condition is generally satisfied in poly(3-alkylthiophenes) when the alkyl chain is heptyl or longer, for which the glass transition is significantly below room temperature.⁵⁰ We hypothesize that unsaturated van der Waals bonds and increased mobility of polymer chains at fractured interfaces, however, may produce skin-depth effects⁸² that facilitate self-healing even in glassy samples.

The self-healing efficiency for polymers intended for structural applications is typically expressed as a fraction of a mechanical property (e.g., strength) of the healed material relative to that of the pristine material.⁷⁶ For self-healing electronic materials, however, an electronic self-healing efficiency must be used alongside a conventional one based on mechanical properties.⁴⁰ Recent research from the laboratory of Kahn and coworkers has shown

that conventional conjugated polymers may already exhibit the capacity for self-repair without any specialized chemical functionality.^{83,84} In particular, laminated interfaces of P3HT showed charge-transport properties that were similar to those of pristine layers of the same thickness.^{83,84}

Conclusions

The design of biologically integrated electronics seems to be a problem for which organic semiconductors are an ideal solution. Despite the potential amenability of organic electronic materials to biological systems, significant work remains to be done to integrate materials from the electronic and biological realms in a way that is seamless. The strategy advocated in this Highlight is the molecular engineering of π -conjugated materials to have properties inspired by biological tissue: extreme elasticity, biodegradability, and the capacity for self-repair. While substantial progress toward this goal has already been made, a concerted, deliberate effort from chemists, materials scientists, engineers, biologists, and the medical community will be required to extract the maximum possible benefit from the versatility of organic semiconducting materials.

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