Li *et al. Soft Sci* 2022;2:7
DOI: 10.20517/ss.2022.07

Soft Science

Perspective

Open Access

Check for updates

PEDOT:PSS-based intrinsically soft and stretchable bioelectronics

Gang Li, Chuan Fei Guo*

Department of Materials Science and Technology, Southern University of Science and Technology, Shenzhen 518055, Guangdong, China.

***Correspondence to:** Prof. Chuan Fei Guo, Department of Materials Science and Technology, Southern University of Science and Technology, 1088 Xueyuan Blvd, Nanshan District, Shenzhen 518055, Guangdong, China. E-mail: guocf@sustech.edu.cn

How to cite this article: Li G, Guo CF. PEDOT:PSS-based intrinsically soft and stretchable bioelectronics. *Soft Sci* 2022;2:7. https://dx.doi.org/10.20517/ss.2022.07

Received: 21 April 2022 **First Decision:** 12 May 2022 **Revised:** 1 June 2022 **Accepted:** 6 June 2022 **Published:** 14 Jun 2022

Academic Editor: Zhifeng Ren **Copy Editor:** Fangling Lan **Production Editor:** Fangling Lan

Abstract

Intrinsically soft and stretchable bioelectronics exhibit tissue-like mechanical behavior that enables the seamless integration of electronic devices with the human body to achieve high-quality biosignal recording and high-efficacy neural modulation. The conducting polymer poly(3,4-ethylenedioxythiophene):polystyrene sulfonate (PEDOT:PSS) shows significant promise in this field because of its high conductivity, excellent biocompatibility and commercial availability. However, pristine PEDOT:PSS is brittle and rigid and thus cannot be used in soft and stretchable electronics. More effort is therefore required to engineer PEDOT:PSS into a stretchable conductor that meets the demands of bioelectronics. In this perspective, we review the recent progress and propose the possible future directions of PEDOT:PSS-based bioelectronics.

Keywords: PEDOT:PSS, bioelectronics, stretchability, electrical conductivity

Bioelectronics play an important role in health monitoring and medical therapies. However, traditional electronic devices are rigid, in contrast to the soft and dynamic nature of biotissues. As a result, rigid bioelectronics suffer from unstable integration with biotissues and may cause a severe immune response when implanted in the human body. A potential solution to this problem relies on endowing electronics with more tissue-like characteristics, such as tissue-like softness and stretchability[1].

Conducting polymers are electronic conductors that exhibit much lower Young's moduli compared with metals, thereby serving as ideal substitutes for metals for soft bioelectronics. Among the various conducting polymers, poly(3,4-ethylenedioxythiophene):polystyrene sulfonate (PEDOT:PSS) [Figure 1A] has attracted significant interest in this field because of its tunable conductivity, excellent biocompatibility and commercial availability[2].

Pristine PEDOT:PSS films made from a commercially available solution, such as Clevios™ PH 1000, often have low conductivity (< 1 S cm^{-1}) and poor stretchability (< 5%) that are insufficient for the requirements of bioelectronics. Early studies have revealed that the addition of a second dopant, such as dimethyl sulfoxide or ethylene glycol, or a certain post-treatment, such as acid treatment, can increase the conductivity of PEDOT:PSS profoundly (> 1000 S cm^{-1})[3]. However, the poor stretchability of PEDOT:PSS still hinders its applications where large deformation is encountered. The Guo group provided a method to make brittle PEDOT:PSS films stretchable by transferring the films onto a biaxial prestrained substrate, followed by releasing the prestrain[4]. Through this process, the conducting PEDOT:PSS film can be elongated to 100% strain without a significant change in resistance, enabled by the dense surface wrinkles in the film.

In addition to structural design for improved stretchability, intrinsically stretchable PEDOT:PSS is also under exploration. The Bao group found that certain ionic liquids (ILs) can serve as second dopants to increase the conductivity of PEDOT and also as plasticizers to increase the stretchability of PEDOT:PSS/IL composites [Figure 1B][5], which performed well in various types of stretchable electronics. However, IL-based conductors are not suitable for bioelectronics, because the possible leakage of toxic ILs in the composite presents a safety issue. Bao and co-workers recently overcame this leakage problem by blending PEDOT:PSS with another polymer containing sliding molecular structures[6]. They claimed that the sliding rings can suppress the crystallinity of polyethylene glycol and thus enhance the stretchability of the blend [Figure 1C]. Through an acid post-treatment, the polymer blend can obtain high conductivity along with high stretchability. Because no small molecules are involved, the polymer blend can be safely implanted in rat models to record neural signals. This work represents a significant milestone in the development of intrinsically soft and stretchable bioelectronics. Furthermore, extra functionalities enrich the potential applications of PEDOT:PSS-based bioelectronics. For instance, Tan *et al.* applied a supramolecular solvent as a dopant to endow a PEDOT:PSS composite with high conductivity and a self-adhesive ability[7]. They used such a composite for bioadhesive electronic devices, which performed stably in monitoring human body activities during exercise. The adhesive property of the PEDOT:PSS composite plays a vital role in signal monitoring because it provides a robust skin-device interface.

Furthermore, fabricating PEDOT:PSS-based hydrogels is another route to intrinsically soft and stretchable bioelectronics. Unlike polymer composites, PEDOT:PSS hydrogels contain a large amount of water, similar to biotissues. Although the incubation of water in PEDOT:PSS-based hydrogels can enhance their biocompatibility with biotissues, it simultaneously brings an even larger challenge. Early studies on conducting polymer hydrogels were focused on the *in-situ* polymerization of the conducting network inside another insulating network. Although these hydrogels may have the desired mechanical properties, their low conductivity (< 0.1 S cm^{-1}) hinders their applications[1]. In 2019, the Zhao group first reported a pure PEDOT:PSS hydrogel, with a single PEDOT:PSS network [Figure 1D], via an annealing treatment[8]. The hydrogel not only exhibited a high conductivity of 40 S cm^{-1} but also enhanced the charge storage/injection capacity compared with traditional metal electrodes. Later, this group showed that the 3D printing of pure PEDOT:PSS hydrogels is an effective method to produce electrode arrays for PEDOT:PSS-based hydrogel bioelectronics [Figure 1E and F][9]. The Bao group fabricated another single network PEDOT:PSS hydrogel by IL-induced gelation and subsequent water exchange[10]. The hydrogel was highly conducting (electrical

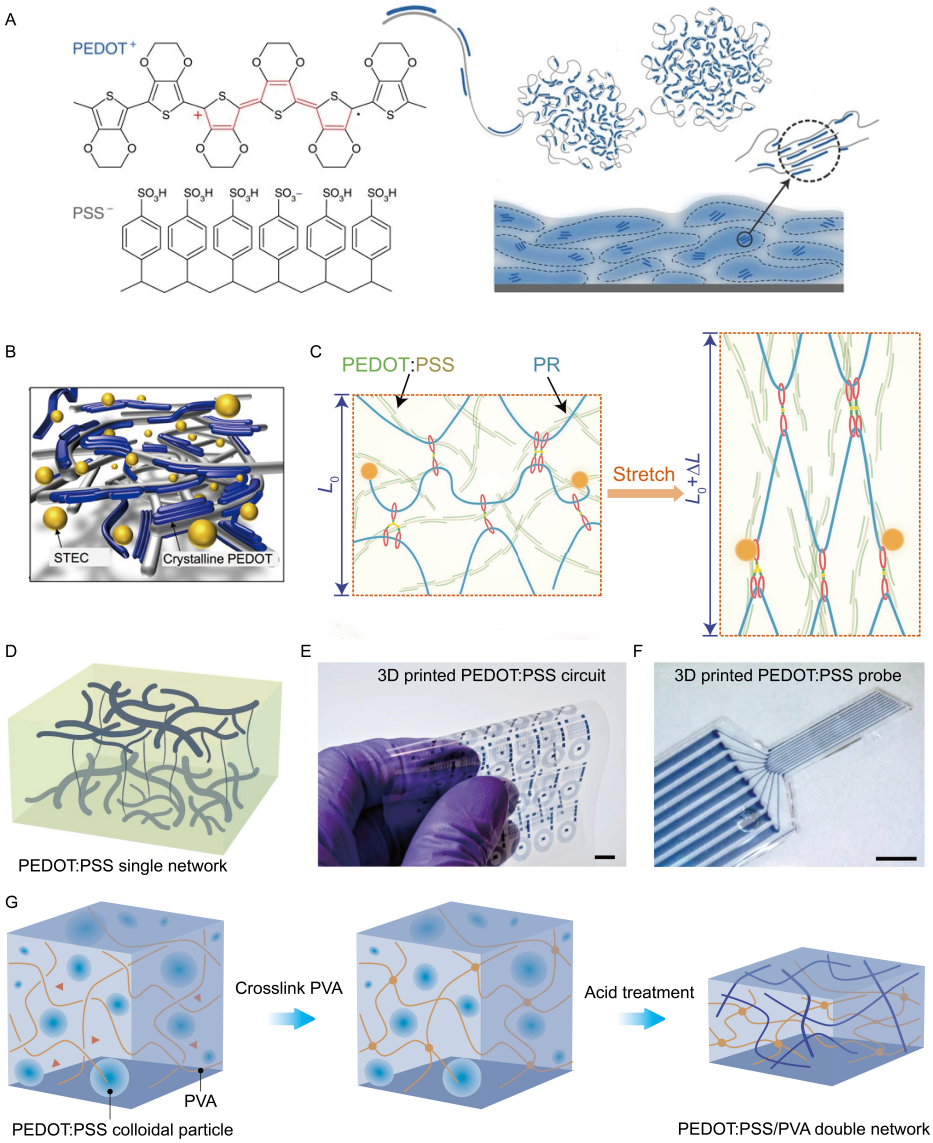

Figure 1. PEDOT:PSS-based intrinsically soft and stretchable bioelectronics. (A) Chemical structure of PEDOT:PSS, the colloidal particle structure of PEDOT:PSS in water dispersion and the microstructure of a PEDOT:PSS film[2]. Reprinted with permission. Copyright 2016, Springer Nature. (B) Schematic illustration of PEDOT:PSS/IL composite, where IL serves as a enhancer of stretchability and electrical conductivity[5]. Reprinted with permission. Copyright 2017, AAAS. (C) Structure of stretchable PEDOT:PSS composite with a topological sliding network[6]. Reprinted with permission. Copyright 2022, AAAS. (D) Pure PEDOT:PSS hydrogel with a single network[8]. Reprinted with permission. Copyright 2019, Springer Nature. Three-dimensional printing of PEDOT:PSS into (E) circuit pattern or (F) neural probe (scale bars of 5 and 1 mm, respectively)[9]. Reprinted with permission. Copyright 2020, Springer Nature. (G) Schematic diagram of PEDOT:PSS/PVA double network hydrogels obtained from a mixed solution to form the PVA network and finally a double network[11]. Reprinted with permission. Copyright 2022, Wiley-VCH.

conductivity of ~47 S cm^{-1}) and could be patterned using photolithography. A bioelectronic device based on this hydrogel was fabricated and implanted in an *in vivo* animal model and showed mild immune response, indicating the significant advantages of hydrogel bioelectronics.

These single network hydrogels are all excellent soft conductors, but their stretchability is limited (elongation at break < 35%). The Guo group resolved this problem using a de-swelling and constrained

aggregation method[11], starting from dispersing PEDOT:PSS colloidal particles in a poly(vinyl alcohol) (PVA) network, then introducing a poor solvent acetic acid to de-swell the PVA network and induce the aggregation of PEDOT:PSS [Figure 1G]. The material eventually becomes a double network hydrogel that exhibits unprecedented balance in electrical conductivity (10 S cm^{-1}) and mechanical stretchability (~150%).

Although PEDOT:PSS-based hydrogels can achieve high conductivity and large stretchability, substantial challenges still remain. First, PEDOT:PSS-based hydrogels not only need to be stretchable, but also fatigue-free over loading-unloading cycles to enable their applications in harsh mechanical conditions. Recently, the Liu group reported a fatigue-resistant hydrogel based on the design of hierarchically aligned structures and well-defined crystalline domains, which showed a high fatigue threshold of 2740 J m^{-2}[12]. Although this hydrogel was non-conducting, the design principle can inspire the exploration of anti-fatigue PEDOT:PSS-based hydrogels. Second, the hydrogels need to be adhesive on wet and dynamic biotissues. Glues or adhesives are required to provide robust and rapid integration of bioelectronic devices on tissues *in vivo,* and safe, on-demand detachment to release the device from the tissue surface is desired. Third, is the softness of the hydrogel programmable? High softness allows for conformable and safer implantation, but only hard devices can penetrate tissues to reach the target site in minor invasive implantations. Can we program the softness of the PEDOT:PSS-based bioelectronics, which is hard during the surgery and soft after implantation? This is an interesting topic for the real applications of PEDOT:PSS-based functional electronics.

DECLARATIONS

Authors' contributions

Wrote the original draft: Li G
Supervised, reviewed and revised the manuscript: Guo CF

Availability of data and materials

Not applicable.

Financial support and sponsorship

Chuan Fei Guo is grateful for the support from the National Natural Science Foundation of China (No. 52073138), the "Guangdong Innovative and Entrepreneurial Research Team Program" under Contract No. 2016ZT06G587, the "Science Technology and Innovation Committee of Shenzhen Municipality" (Grant No. JCYJ20210324120202007), and the Shenzhen Sci-Tech Fund (No. KYTDPT20181011104007).

Conflicts of interest

Both authors declared that there are no conflicts of interest.

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

REFERENCES

1. Yuk H, Lu B, Zhao X. Hydrogel bioelectronics. *Chem Soc Rev* 2019;48:1642-67. DOI PubMed
2. Rivnay J, Inal S, Collins BA, et al. Structural control of mixed ionic and electronic transport in conducting polymers. *Nat Commun* 2016;7:11287. DOI PubMed PMC
3. Shi H, Liu C, Jiang Q, Xu J. Effective approaches to improve the electrical conductivity of PEDOT:PSS: a review. *Adv Electron Mater* 2015;1:1500017. DOI
4. Li G, Qiu Z, Wang Y, et al. PEDOT:PSS/Grafted-PDMS electrodes for fully organic and intrinsically stretchable skin-like electronics. *ACS Appl Mater Interfaces* 2019;11:10373-9. DOI PubMed
5. Wang Y, Zhu C, Pfattner R, et al. A highly stretchable, transparent, and conductive polymer. *Sci Adv* 2017;3:e1602076. DOI PubMed PMC
6. Jiang Y, Zhang Z, Wang YX, et al. Topological supramolecular network enabled high-conductivity, stretchable organic bioelectronics. *Science* 2022;375:1411-7. DOI PubMed
7. Tan P, Wang H, Xiao F, et al. Solution-processable, soft, self-adhesive, and conductive polymer composites for soft electronics. *Nat Commun* 2022;13:358. DOI PubMed PMC
8. Lu B, Yuk H, Lin S, et al. Pure PEDOT:PSS hydrogels. *Nat Commun* 2019;10:1043. DOI PubMed PMC
9. Yuk H, Lu B, Lin S, et al. 3D printing of conducting polymers. *Nat Commun* 2020;11:1604. DOI PubMed PMC
10. Liu Y, Liu J, Chen S, et al. Soft and elastic hydrogel-based microelectronics for localized low-voltage neuromodulation. *Nat Biomed Eng* 2019;3:58-68. DOI PubMed
11. Li G, Huang K, Deng J, et al. Highly conducting and stretchable double-network hydrogel for soft bioelectronics. *Adv Mater* 2022;34:e2200261. DOI PubMed
12. Liang X, Chen G, Lin S, et al. Anisotropically fatigue-resistant hydrogels. *Adv Mater* 2021;33:e2102011. DOI PubMed