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Synthesis of Organic Semiconducting Thin Films and Their Prospects in Flexible Sensor Technologies

Abstract

This study investigates the synthesis of pure hydrogel films based on the organic semiconductor material PEDOT:PSS and their potential application in flexible sensor devices. Through a controlled drying and rehydration process facilitated by the addition of dimethyl sulfoxide (DMSO), the resulting films exhibit high electrical conductivity (~20–40 S/cm), excellent elasticity (>35%), and strong biocompatibility. These films also demonstrate a low Young's modulus (~2 MPa) and high water content, ensuring mechanical durability in physiological environments. These combined properties make PEDOT:PSS hydrogel films promising candidates for next-generation flexible and biointegrated sensor technologies.

Keywords: *semiconductors, synthesis, sensors, flexibility, polymers*

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Üzvi yarımkeçirici nazik təbəqələrin sintezi və onların perspektivləri çevik sensor texnologiyalarında

Xülasə

Bu tədqiqat, üzvi yarımkeçirici material olan PEDOT:PSS əsasında saf hidrogel təbəqələrinin sintezini və onların çevik sensor qurğularında tətbiq potensialını araşdırır. Dimetilsulfoksid (DMSO) əlavəsi ilə aparılan nəzarətli qurudulma və rehidrasiya prosesi nəticəsində əldə edilən təbəqələr yüksək elektrik keçiriciliyi (~20–40 S/sm), əla elastiklik (>35%) və güclü biouyğunluq nümayiş etdirir. Bu təbəqələr, həmçinin aşağı Yununq modulu (~2 MPa) və yüksək su tərkibi ilə seçilərək fizioloji mühitdə mexaniki davamlılığı təmin edir. Bu xüsusiyyətlərin birgə təsiri PEDOT:PSS əsaslı hidrogel təbəqələrini növbəti nəsil çevik və biointeqrasiya olunmuş sensor texnologiyaları üçün perspektivli namizədə çevirir.

Açar sözlər: *yarımkeçiricilər, sintez, sensorlar, elastiklik, polimerlər*

Introduction

In the 21st century, one of the main frontiers of scientific and technological progress is the advancement of flexible and wearable electronic devices. Developments in this area have enabled a broad spectrum of applications, including medical diagnostics, human-machine interfaces, artificial skin, environmental sensing and bioelectronic platforms. For these systems to function effectively and consistently, the materials involved must combine excellent electrical conductivity with mechanical flexibility. Conventional inorganic semiconductors—such as silicon (Si), gallium arsenide (GaAs) and indium tin oxide (ITO)—fall short of these requirements due to their rigid and brittle nature and limited biocompatibility. In response, organic semiconductors, particularly conjugated polymers like PEDOT:PSS (poly(3,4-ethylenedioxythiophene):polystyrene sulfonate) and polyaniline, have gained prominence as viable alternatives (Liu, 2012; Chen et al., 2003). Their conjugated structures facilitate charge delocalization, contributing to high conductivity, while their inherent flexibility supports the development of durable, bendable thin films (Li et al., 2023). Additionally, their ability to be processed in water-based solutions, fabricated at low temperatures, and integrated into transparent electronic systems enhances their suitability for modern manufacturing techniques. Their compatibility with biological systems further makes them ideal for use in implantable devices and neural interfaces that require direct interaction with living tissues (Yue et al., 2011; Bell, 2008).

Despite these advantages, producing high-performance thin films from organic semiconductors poses significant challenges. Issues such as suboptimal conductivity, structural defects, limited mechanical strength, and long-term instability persist. To mitigate these drawbacks, researchers have explored methods including chemical doping, molecular design modifications, and optimized drying techniques (He et al., 2013; Bubnova et al., 2012). A particularly effective strategy for enhancing PEDOT:PSS performance involves the use of polar solvents—especially dimethyl sulfoxide (DMSO)—to create hydrogel-based pure thin films. This technique allows for the development of highly conductive, flexible, and water-retentive structures (Tritt et al., 2006). In this study, dimethyl sulfoxide (DMSO), a polar co-solvent with a high boiling point, was incorporated into an aqueous PEDOT:PSS solution as a functional additive. DMSO facilitates the recrystallization of PEDOT-rich nanofibrils and promotes the reorganization of PEDOT:PSS polymer chains during the dry-annealing phase. Crucially, both water and DMSO are entirely eliminated through an extensive drying process (24 hours at 60 °C), followed by three annealing cycles at 130 °C for 30 minutes each. This processing method leads to the formation of pure PEDOT:PSS hydrogel films. Notably, (Chen et al., 2003) introduced an innovative approach based on sequential drying and rehydration, producing films that are not only mechanically soft and compliant but also maintain electrical stability. These films exhibited favorable characteristics such as a low Young's modulus (~2 MPa), high water content (>80%), biocompatibility, and sustained structural robustness—traits that render them well-suited for emerging biointegrated sensor technologies (Venkatasubramanian et al.,). The present research investigates the synthesis of such materials, assesses their physicochemical and electrical characteristics, and explores their suitability for use in flexible sensor devices. The main goal is to evaluate the potential of PEDOT:PSS-based thin films as functional components in advanced sensor systems and to contribute foundational insights for their continued development and practical deployment (Dubey et al.; Bubnova et al., 2011).

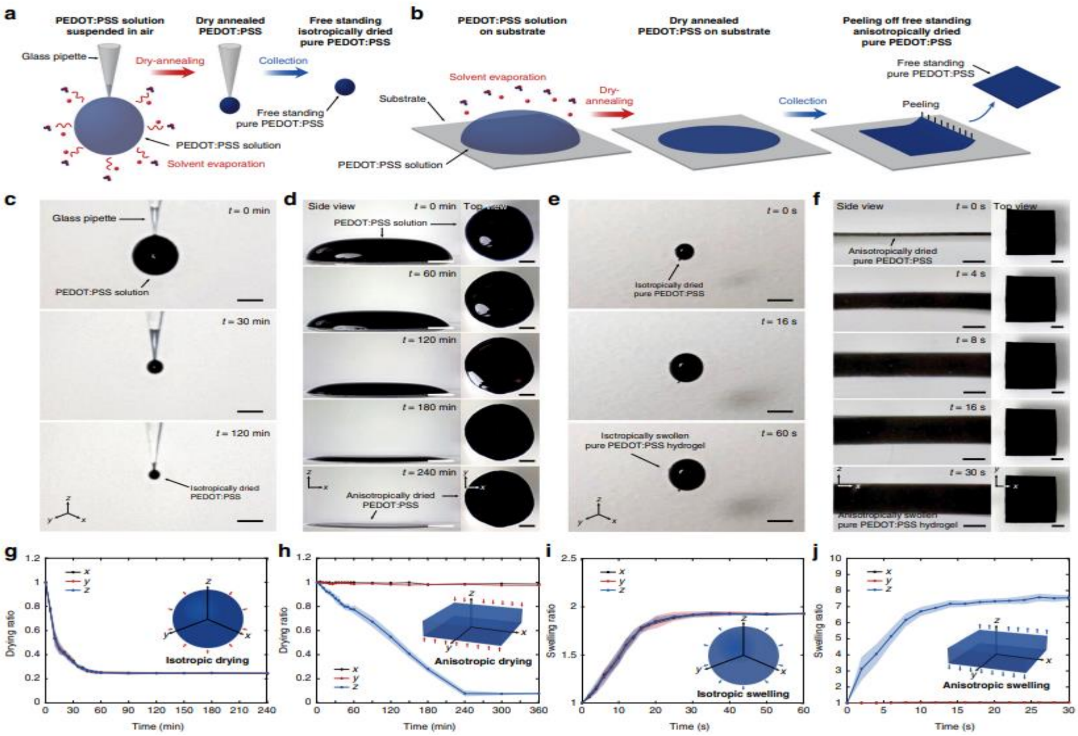

Fig. 1. Dry-Annealing and Swelling Characteristics of Pure PEDOT:PSS Hydrogels.
a. Schematic depiction of isotropic dry-annealing of aqueous PEDOT:PSS solution resulting in a microball formation.
b. Schematic representation of anisotropic dry-annealing producing a free-standing film from aqueous PEDOT:PSS solution.
c. Experimental images showing the isotropic dry-annealing process leading to the formation of a microball.
d. Experimental images demonstrating anisotropic dry-annealing that yields a free-standing film.
e. Isotropic swelling behavior of a dry-annealed PEDOT:PSS microball transitioning into a stable hydrogel state.
f. Anisotropic swelling of a free-standing PEDOT:PSS film into a structurally stable hydrogel.
g, h. Time-dependent dimensional variations observed during g isotropic and h anisotropic dry-annealing of aqueous PEDOT:PSS.
i, j. Time-course analysis of dimensional changes during i isotropic and j anisotropic swelling of the resulting pure PEDOT:PSS hydrogels. Data in panels g–j are presented as means, with error bars indicating standard deviation (n = 3). Scale bars represent 1 mm.

Research

This study aims to fabricate pure hydrogel thin films based on PEDOT:PSS that exhibit both high electrical conductivity and mechanical durability, while also evaluating their suitability for use in flexible bioelectronic sensors (Yue R., Xu J.,2012), (Poehler T. O., Katz & H. E., 2012). To modify the properties of the PEDOT:PSS solution, varying concentrations of dimethyl sulfoxide (DMSO) were introduced. The prepared samples underwent a three-stage drying process: an initial 24-hour drying period at 60 °C, followed by three annealing cycles of 30 minutes each at 130 °C. After drying, the films were rehydrated to form hydrogel structures. The final hydrogel films were then analyzed to assess their electrical conductivity, mechanical flexibility, and structural characteristics.

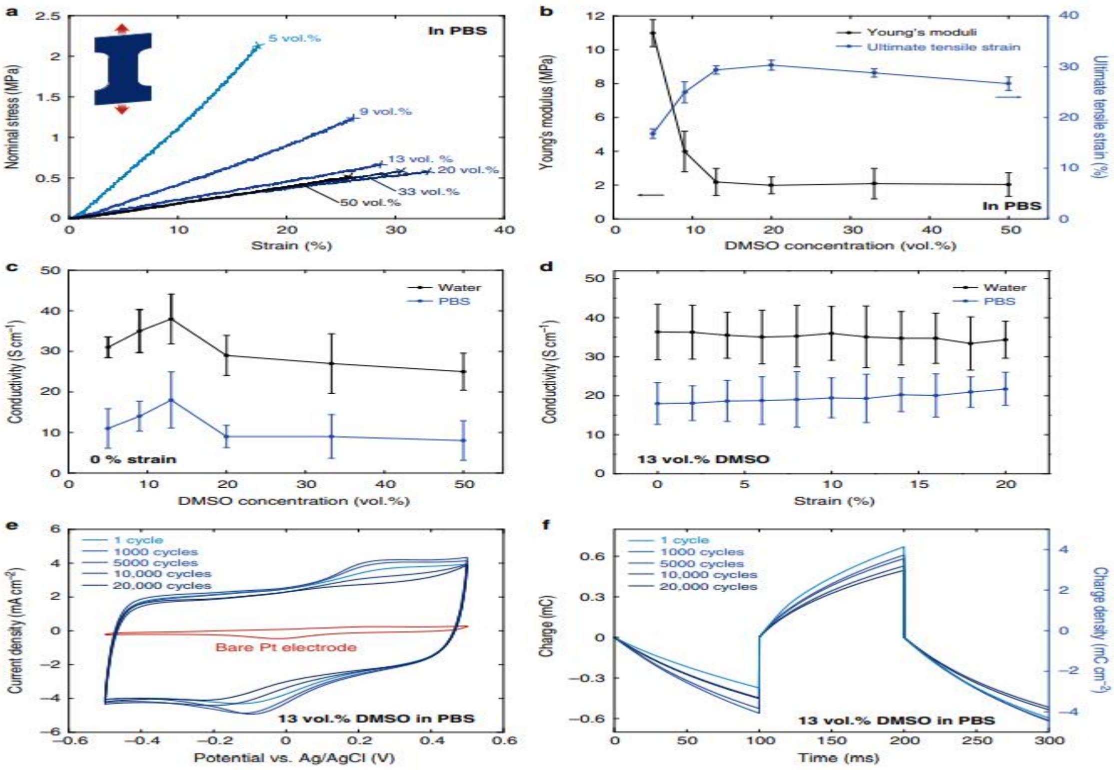

Fig. 2. Mechanical and Electrical Properties of Pure PEDOT:PSS Hydrogels

a. Representative nominal stress–strain curves for free-standing PEDOT:PSS hydrogels immersed in PBS, measured at different concentrations of DMSO.

b. Variation in Young's modulus and maximum tensile strain of the hydrogels as a function of DMSO concentration in PBS.

c. Electrical conductivity of PEDOT:PSS hydrogels measured in both deionized water and PBS across a range of DMSO concentrations.

d. Conductivity profiles of the hydrogels under different applied strains, evaluated in deionized water and PBS.

e. Cyclic voltammetry (CV) curves of the PEDOT:PSS hydrogel-coated platinum (Pt) electrode in PBS. For comparison, the CV curve of an uncoated (bare) Pt electrode is shown in red.

f. Cyclic current response of the PEDOT:PSS hydrogel-coated Pt electrode under repetitive voltage pulses ranging from −1.5 V to +1.5 V (vs. Ag/AgCl reference).

Data in panels **b–d** are reported as mean values, with error bars representing standard deviations (n = 4).

Table 1.
Properties of PEDOT:PSS films according to DMSO concentration.

DMSO (%)	Conductivity (S/cm, PBS)	Conductivity (S/cm, water)	Young's Modulus (MPa)	Strain at Break (%)
5	~8	~14	10	20
13	~20	~40	2	35
20	~18	~36	3	32
33	~10	~25	4	28
50	~5	~10	5	22

The best performance was observed when the DMSO concentration reached 13%. Under these conditions, the resulting films achieved electrical conductivities of approximately 20 S/cm in phosphate-buffered saline (PBS) and about 40 S/cm in water. In terms of mechanical properties, the films showed remarkable elasticity, with a Young's modulus of 2 MPa and a strain at break of 35%, indicating strong compatibility with soft biological tissues. Notably, the films preserved both their conductivity and structural integrity for up to three months, demonstrating excellent long-term stability. Additionally, variations in swelling behavior between isotropic and anisotropic drying techniques—such as calibration and template printing—further support the versatility of these films in different sensor manufacturing processes. Overall, this study marks a significant step forward in developing PEDOT:PSS-based hydrogel thin films that combine superior conductivity with mechanical resilience, confirming their potential for integration into flexible bioelectronic sensors. The results offer important perspectives for advancing research in material science, biotechnology, and sensor technologies.

Conclusion

This study has shown that pure PEDOT:PSS-based hydrogel films hold significant promise as organic semiconductors for use in flexible sensor technologies. The incorporation of dimethyl sulfoxide (DMSO) during the synthesis process greatly improves the films' electrical conductivity, mechanical flexibility, and stability in aqueous environments. These enhanced characteristics make PEDOT:PSS hydrogels particularly well-suited for practical applications in implantable bioelectronics, skin-mounted sensors, and neural interface devices. Their high elasticity allows them to closely mimic the mechanical behavior of human skin and tissues, improving comfort and reducing the risk of tissue irritation or damage during prolonged use. Moreover, their water resistance ensures consistent performance in humid and physiologically relevant conditions, which is essential for dependable biomedical sensing. Future investigations should aim to produce these materials in various micro- and nanoscale structures by leveraging advanced manufacturing techniques such as inkjet and 3D printing, enabling the creation of highly precise and customizable sensors. Additionally, chemical functionalization of PEDOT:PSS hydrogels could further enhance their biocompatibility, molecular selectivity, and responsiveness to specific biological stimuli. These innovations would support the development of more sophisticated and multifunctional bioelectronic platforms. In conclusion, PEDOT:PSS-based hydrogel films represent a highly promising class of materials for the next generation of flexible, high-performance, and long-lasting sensor systems. This research also contributes to the refinement of scalable and efficient fabrication approaches for organic semiconductors, opening up new possibilities for their broader industrial adoption.

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