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## Research Article

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# Solvothermal Engineering: A Universal Route to High-Performance Free-standing PEDOT:PSS Thick Films

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**Abstract:** Electrical conductivity is a critical parameter for conductive polymers. Conventional strategies of solvent-doping and solvent post-treatment have improved the conductivity of pure free-standing PEDOT:PSS films from  $\sim 2$  to  $\sim 1500 \text{ S cm}^{-1}$ , yet substantial room for improvement remains. The solvothermal technique, with its unique advantage of high temperature and self-generated pressure, demonstrates significant potential for further enhancing the conductivity of free-standing PEDOT:PSS thick films. Herein, we systematically investigated the solvothermal effects of methanol (MeOH), N, N-dimethylformamide (DMF), and dimethyl sulfoxide (DMSO) on the conductivity of free-standing PEDOT:PSS thick films. The obtained free-standing PEDOT:PSS films exhibited high conductivities of  $2958 \text{ S cm}^{-1}$  (MeOH),  $2528 \text{ S cm}^{-1}$  (DMF), and  $2438 \text{ S cm}^{-1}$  (DMSO), respectively. This exceptional performance arise from the synergistic effect of enhanced  $\pi$ - $\pi$  stacking, increased crystallinity and aggregation of PEDOT nanocrystals, the transition of PEDOT chains from a neutral state to a bipolaronic state, structural transformation of PEDOT from benzene to quinone forms, and massive PSS removal. What excites us is that the PEDOT film achieved a capacitance of  $251 \text{ F cm}^{-3}$  due to the optimization of the preparation process, significantly surpassing previously reported values. This study verifies the universality of solvothermal strategy with common organic solvents for improving the conductivity

of PEDOT:PSS thick film and highlights its great promise for energy storage field.

**Keywords:** PEDOT:PSS, Solvothermal Engineering, Organic Solvents, Conductivity, Capacitance

## 1. Introduction

Poly (3,4-ethyl dioxythiophene):Poly (styrene sulfonic acid) (PEDOT:PSS), a conductive polymer, is highly regarded in commercial community for its tunable conductivity, excellent thermal stability and broad processability.[1,2] It has been extensively applied in energy-related devices[3,4], including solar cells[3], thermoelectric devices[5,6], and supercapacitors (SCs)[7,8]. The current reported PEDOT:PSS films could be classified into thin film (nanometer level, nm) and thick film (micrometer level,  $\mu\text{m}$ ). Thick films offer an increased number of charge carrier pathways, making them well-suited for high-current applications. Meanwhile, their inherent self-supporting nature facilitates integration into wearable electronic systems. PEDOT:PSS films serve as a class of conductive films, in which conductivity is a critical parameter. As an alternative electrode, high conductivity of PEDOT:PSS thick film makes it possible to replace conventional electrode materials, such as indium tin oxide (ITO), which exhibits conductivity exceeding  $3000\text{ S cm}^{-1}$ . In thermoelectric applications, high conductivity directly contributes to the thermoelectric conversion efficiency. In SCs, high conductivity enables faster charge transport, allowing the device to charge and discharge more quickly, thereby enhancing power density and rate capability.

Many studies have been devoted to improving the conductivity of PEDOT:PSS. The strategies could be classified into physical and chemical approaches. Physical methods, including annealing, light-induced deformation, and mechanical stretching, were employed to enhance conductivity by improving crystallization and molecular alignment of PEDOT:PSS.[9] These techniques typically function by disrupting the interactions between PEDOT and PSS chain. For example, Kim et al. reported an increase in the conductivity of PEDOT:PSS film to  $0.2\text{ S cm}^{-1}$  by adjusting annealing temperature between 100-200 °C.[10] Chemical techniques primarily involve treating

PEDOT:PSS film with solvents, including organic solvents, ionic liquids, and acids. The solvent-treatment methods can be classified into two categories: solvent addition (during solution preparation) and solvent post-treatment (after film formation).[11,12] Among the various solvents, polar solvents with diverse boiling points are usually used due to the ability to destroy the electrostatic interaction between PEDOT and PSS, promoting conformational transitions in PEDOT chains, and optimizing phase separation and film morphology. For example, Saxena et al. treated PEDOT:PSS films with 1-ethyl-3-methylimidazole tetracyanoborate (EMIM TCB) to realize a conductivity of  $600 \text{ S cm}^{-1}$ . [13] In contrast, Badre et al. mixed EMIM TCB directly with the PEDOT:PSS solution to prepare a transparent film with an enhanced conductivity of  $2084 \text{ S cm}^{-1}$ . [14] Zhang et al. treated PEDOT:PSS films with 80% N, N-dimethylformamide (DMF) and 20% water, resulting in a conductivity of  $1055 \text{ S cm}^{-1}$ . [15]

In general, solvent post-treatment is more effective than solvent addition in enhancing the conductivity of PEDOT:PSS films.[16] However, as the film thickness increases, the effectiveness significantly diminishes, because the solvents encounter extremely large challenges in the process of penetrating into the interior of the thick film. Currently, the conductivity of pure PEDOT:PSS (PH1000) thick film is typically limited to  $\sim 1000 \text{ S cm}^{-1}$  with traditional acid-free solvent treatment. The solvothermal method, as an extension of hydrothermal technology, could simultaneously offer high temperature and self-generated pressure within a sealed system and thus provides a significant enhancement to the solvent's penetration ability. Meanwhile, the high-temperature and high-pressure environment could simultaneously enhance the arrangement of PEDOT molecules, thereby improving electrical performance.[17] During the solvothermal process, the penetration of the solvent effectively removes excess PSS, exposes more PEDOT chains, and significantly enhances conductivity.[18] In addition, polar solvents promote conformational rearrangement of PEDOT, which can also contribute to increased capacitance. In our previous studies, we firstly developed a solvothermal method for processing thick PEDOT:PSS films using concentrated sulfuric acid, a high conductivity of  $3188 \text{ S cm}^{-1}$  and a moderate

capacitance of  $110 \text{ F cm}^{-3}$  were achieved.[19] This study highlights the great potential of solvothermal techniques for improving the conductivity and capacitance of PEDOT:PSS. Yet, the strong corrosiveness of concentrated sulfuric acid makes it difficult to precisely control the morphology of PEDOT:PSS and thus achieve a high performance, and it also poses significant safety risks. To date, a systematic investigation of solvothermal treatment with common solvents for PEDOT:PSS films is still lacking.

Methanol (MeOH), dimethyl sulfoxide (DMSO) and N, N-dimethylformamide (DMF) were chosen after considering their boiling points and polarity. The detailed parameters of boiling points, polarity, and Hansen solubility parameters are shown in **Table S1**. These gradient physicochemical differences govern the PSS removal efficiency, thereby effectively tuning the molecular conformation, microstructure, and electrical and capacitive performance of the treated PEDOT:PSS films.

Herein, we employed mild solvents to treat PEDOT:PSS thick films with solvothermal technology and systematically explored the effects on the electrical performance. We systematically studied the effects of different organic solvents on PEDOT arrangement and molecular structure, film morphology and electrochemical properties by employing a variety of different characterization methods. The results demonstrate that the electrical conductivity of the treated PEDOT:PSS thick films exceeded  $2400 \text{ S cm}^{-1}$ , confirming the broad universality and effectiveness of the solvothermal method across a range of commonly used organic solvents. Among them, the conductivity of PEDOT:PSS films treated with MeOH reached as high as  $2958 \text{ S cm}^{-1}$ , and the capacitance performance can be enhanced to  $180 \text{ F cm}^{-3}$ , surpassing all previously reported values for MeOH-based treatments, including doping, post-treatment, and immersion approaches.[20-23] These findings highlight the superior potential of the solvothermal method in enhancing conductivity and capacitance of conducting polymers. This work provides an effective strategy for improving the performance of PEDOT:PSS thick films and it will promote their applications of PEDOT:PSS in the field of SCs.

## **2. Experimental Section**

### **2.1 Materials**

Aqueous PEDOT:PSS (Clevios PH1000, weight ratio: PEDOT:PSS=1:2.5) was purchased from Xi'an Polymer Light Technology Corporation. Methanol (MeOH) (99.9 wt%), dimethyl Sulfoxide (DMSO) (99.7 wt%), and N,N-Dimethylformamide (DMF) (99 wt%) were purchased from Anhui Zesheng Technology Co. Ltd. The nano-porous polypropylene membranes (Celgard 3501) with a pore size of 0.064  $\mu\text{m}$  were supplied by Celgard LLC.

### **2.2 Preparation of free-standing PEDOT:PSS films**

Firstly, the nano-porous polypropylene membrane was placed on the rotary table of a vacuum filtration apparatus, and 1 mL PH1000 solution was vacuum-filtered onto the membrane under ambient conditions for 3 h to form PEDOT:PSS films. Subsequently, the films were dried at 25 °C in air for 6 h. After drying, the films were peeled off the polypropylenes membrane to obtain free-standing PEDOT:PSS films.

### **2.3 Post-treatment of free-standing PEDOT:PSS films by solvothermal methods**

The free-standing films were transferred into a 100 mL Teflon-lined stainless-steel autoclave containing 50 mL of solvent (MeOH, DMF, or DMSO) and subjected to solvothermal treatment at various temperatures for 6 h. For MeOH, the films were heated at 30 °C, 50 °C, 60 °C, and 80 °C, and the resulting samples were abbreviated as MeOH-30, MeOH-50, MeOH-60, and MeOH-80. For DMSO, the treatment temperatures were set at 80 °C, 100 °C, 120 °C, and 140 °C, with the corresponding samples labeled as DMSO-80, DMSO-100, DMSO-120, and DMSO-140. Similarly, for the DMF, the films were treated at 80 °C, 100 °C, 120 °C, and 140 °C, and named as DMF-80, DMF-100, DMF-120, and DMF-140. The reaction pressures were presented in **Tables S2, S3, and S4**. After the solvothermal treatment, the autoclave was allowed to cool naturally to room temperature, the films were then retrieved from the autoclave, rinsed with deionized water to remove residual solvent residues, and subsequently dried under ambient conditions.

## 2.4 Characteristics

The thickness of free-standing PEDOT:PSS films was measured using a Dektak-XT Brooke (Bruker) step instrument. Structural characterization was performed by X-ray photoelectron spectroscopy (XPS, K-Alpha, Thermo Scientific, USA) using Al K $\alpha$  rays at 12 kV. Raman spectroscopy was performed on the LabRAMHR800 (Horiba Jobin Yvon). The Ultraviolet-visible (UV-vis) spectra of the samples were recorded with a UV-vis spectrometer (UV-vis, Shimadzu UV-3600i Plus, Japan). Electron spin resonance spectroscopy (ESR/EPR) was carried out on a Bruker EMXplus-6/1 (Germany). Grazing-incidence wide-angle X-ray scattering (GIWAXS) was performed using a Xenocs (XeuSS) with a photon wavelength of 1.54 Å. PILATUS3 R 300K area detector was used and positioned 130 mm from the sample center. Conductive atomic force microscopy (EFM) was conducted using a Jupiter XR scanning probe microscope equipped with a conductive probe (ASYELEC-01-R2). Height and current images were collected in contact mode under a bias voltage of 0.2 V. The sheet resistance of the films was measured using a four-point probe technique (RTS-8, Guangzhou, China).

Electrochemical performance, including cyclic voltammetry (CV) and galvanostatic charge-discharge measurements (GCD) was measured using an electrochemical workstation (CHI660E, CH Instruments, Shanghai, China). Volume capacitance of one single electrode was calculated according to the following equation:  $C = I \Delta t / \Delta E$ , or  $C_v = C/V = I \Delta t / V \Delta E$ , where  $C$  is the total capacitance,  $C_v$  is the specific capacitance in  $\text{mF cm}^{-3}$ ,  $I$  is the discharge current in  $\text{A cm}^{-3}$ ,  $\Delta t$  is the discharge time in s,  $\Delta E$  is the potential window in the discharge process after the current drops in V, and  $V$  is the volume of the PEDOT:PSS films in  $\text{cm}^{-3}$ .

## 3. Results and Discussion

### 3.1 Conductivity

Pristine PEDOT:PSS thick films were treated by solvothermal processing using MeOH, DMF, and DMSO acid-free solvents. The corresponding thicknesses, resistances, and conductivity of the above PEDOT:PSS films are presented in **Tables S5-S7**. The correlation plot of pressure vs. conductivity was shown in **Figure S1**. The

thicknesses of solvothermal-treated samples decreased slightly from the initial thickness of 4.0  $\mu\text{m}$  for pristine PEDOT:PSS films (PH1000) to 3.6-3.8  $\mu\text{m}$ , demonstrating a denser and more compact film structure. The reduction is attributed to the pressure-induced PSS part removal and molecular rearrangement.[18] Meanwhile, the conductivity of MeOH-treated samples increased with pressure, attributed to the small MeOH molecules readily penetrating into PEDOT:PSS thick films, promoting partial PSS removal and molecular rearrangement. In contrast, DMF- and DMSO-treated samples showed an initial increase followed by a decrease in conductivity, likely due to their larger molecular sizes and higher boiling points. With increasing pressure, these solvents were more likely to be retained within the films, thereby reducing conductivity. As shown in **Figure 1a**, the conductivity of nearly all solvothermal-treated samples exceeded 1500  $\text{S cm}^{-1}$  (**Figure 1a**), surpassing both the untreated PEDOT:PSS films (2  $\text{S cm}^{-1}$ ) and those processed by conventional methods (doping or dipping techniques) using the same solvents (**Figure 1b**). The maximum conductivity with MeOH, DMF, and DMSO solvents could reach 2958  $\text{S cm}^{-1}$  (MeOH-80), 2528  $\text{S cm}^{-1}$  (DMF-100), and 2438  $\text{S cm}^{-1}$  (DMSO-100), respectively. These results demonstrate that solvothermal treatment is more effective and universal for improving the conductivity of PEDOT:PSS thick films, achieving performance improvements not attainable by existing methods.

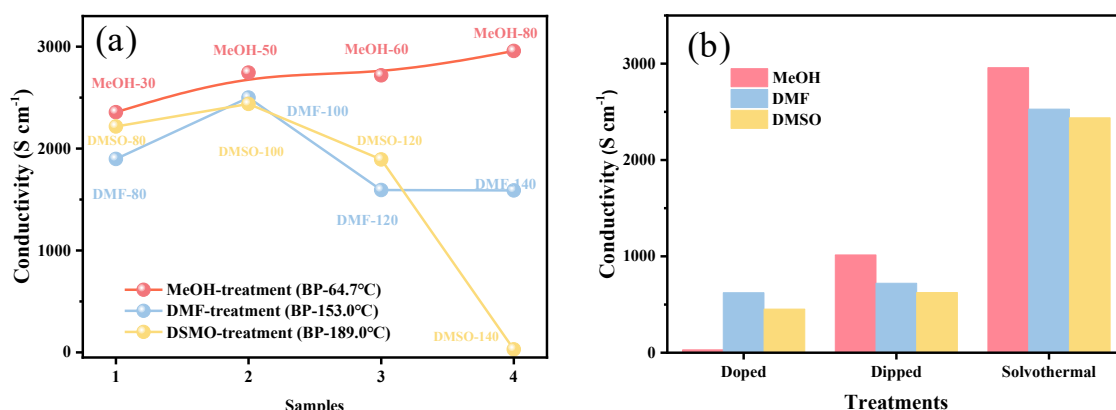


**Figure 1** (a) The conductivity of pristine PEDOT:PSS thick films treated by MeOH, DMF, and DMSO solvents; (b) The conductivity achieved by doping, dipping, and solvothermal approaches with MeOH, DMF, and DMSO solvents.[16, 24, 25]

The remarkable achievements can be attributed to microstructural alterations, stemming from the solvothermal effect. Therefore, the combination of GIWAXS, XPS, Raman spectra, UV-*vis*, ESR/EPR, and CAFM, was employed to characterize the structural evolution of PEDOT:PSS thick films throughout the solvothermal process.

GIWAXS was employed to monitor molecular-level rearrangement of PEDOT:PSS films subjected to different solvent-based solvothermal treatments. The corresponding 2D GIWAXS patterns are shown in **Figure 2a-e**. As shown in **Figure 2a-d**, the treated films exhibit a higher bright intensity along  $q_{xy}$  for  $\pi$ - $\pi$  stacking of the PEDOT chain, compared to the untreated PEDOT:PSS film. The phenomenon indicates a rearrangement of  $\pi$ - $\pi$  stacking into an edge-on orientation, which is beneficial to in-plane conduction.[26] Specifically, the intensity trend follows MeOH-80 > DMF-100  $\approx$  DMSO-100, consistent with the observed conductivity trend of MeOH-80 > DMF-100  $\approx$  DMSO-100. As shown in GIWAXS curves of **Figure 2e**, two peaks at  $q_z=0.308 \text{ \AA}^{-1}$  and  $0.548 \text{ \AA}^{-1}$  along the  $q_z$  direction are observed in the low angle region, representing the first-order (100) and second-order (200) of the PEDOT nanocrystals, respectively.[27] In the high angle region, broad peaks at  $q=1.781 \text{ \AA}^{-1}$  and  $q=1.223 \text{ \AA}^{-1}$  are assigned to the  $\pi$ - $\pi$  stacking of the PEDOT chain in the (010) direction and the nanocrystalline ring of PSS, respectively.[23] Similar peaks were found in the solvothermal-treated PEDOT:PSS thick films, indicating that the fundamental structure of PEDOT:PSS was maintained during solvothermal treatment. Generally, the higher peak intensity indicates greater molecular ordering and crystallinity, contributing directly to improved charge transport.[28] The MeOH-80 sample exhibits the highest crystallinity in the low-angle region, which is consistent with its highest conductivity among all samples. To further quantify structural changes,  $d$ -spacings were calculated from the positions of the (100), (200), and (010) peaks (**Table 1**). Along the (010) direction, the  $d$ -spacing decreases in solvothermal-treated films compared to the untreated film, suggesting stronger  $\pi$ -orbital overlap, which facilitates faster charge carrier transport.[29,30] In addition, the  $d$ -spacings along the (100) and (200) directions slightly increase among the treated samples, suggesting the formation of layered PEDOT:PSS structures (**Figure S2**), possibly due to solvent-induced molecular

rearrangement or partial PSS removal.

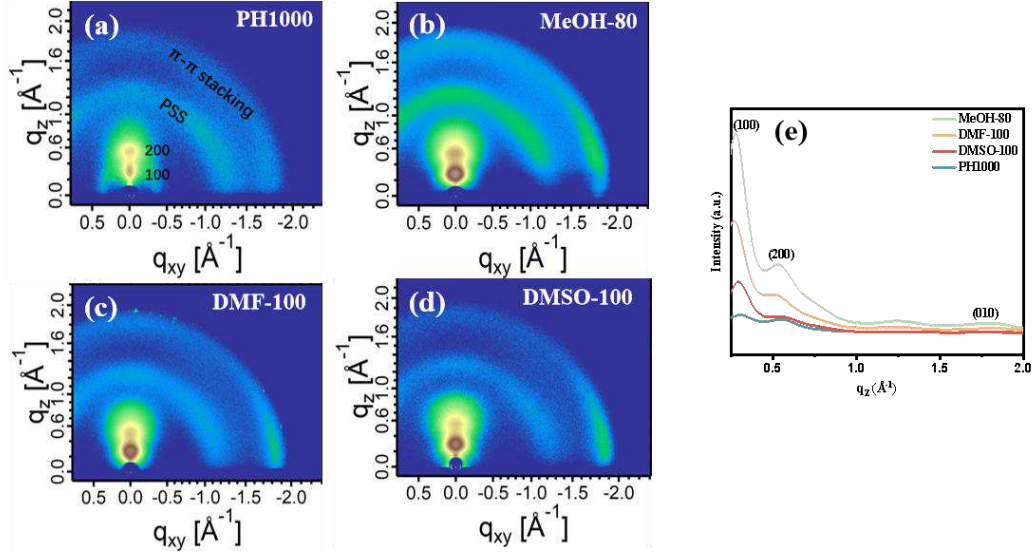


**Figure 2** 2D GIWAXS patterns of (a) PH1000, (b) MeOH-80, (c) DMF-100, and (d) DMSO-100, (e) GIWAXS scattering plots along the  $q_z$  direction.

**Table 1** The location of PEDOT nanocrystals and PSS chains and their  $d$  distances.

| Sample   | PEDOT      |          |            |          | PSS        |          | /     |
|----------|------------|----------|------------|----------|------------|----------|-------|
|          | $q_z(100)$ | $d(100)$ | $q_z(200)$ | $d(200)$ | $q_y(010)$ | $d(010)$ |       |
| PH1000   | 0.308      | 20.390   | 0.548      | 11.460   | 1.781      | 3.526    | 1.223 |
| MeOH-80  | 0.278      | 22.590   | 0.534      | 11.760   | 1.781      | 3.526    | 1.254 |
| DMF-100  | 0.271      | 23.173   | 0.496      | 12.661   | 1.827      | 3.437    | 1.223 |
| DMSO-100 | 0.293      | 21.433   | 0.548      | 11.460   | 1.823      | 3.445    | 1.286 |

**Figure 3a** presents the Raman spectra of PH1000, MeOH-80, DMF-100, and DMSO-100. The peaks at  $1363\text{ cm}^{-1}$  and  $1438\text{ cm}^{-1}$  belong to  $C_\beta$ - $C_\beta$  single bond stretching and  $C_\alpha=C_\beta$  symmetric stretching, respectively.[31] The peaks at  $1494\text{ cm}^{-1}$  and  $1557\text{ cm}^{-1}$  are ascribed to  $C_\alpha=C_\beta$  antisymmetric stretching and PSS vibration, respectively.[32] Compared with the untreated PH1000 film, the solvothermal-treated samples show a pronounced red shift, particularly in the  $C_\alpha=C_\beta$  symmetric stretching region. Those peaks shift from  $1438\text{ cm}^{-1}$  (PH1000) to  $1422\text{ cm}^{-1}$  (MeOH-80),  $1423\text{ cm}^{-1}$  (DMF-100), and  $1430\text{ cm}^{-1}$  (DMSO-100). This red shift indicates a structural transition of the PEDOT chains from the neutral benzene form to a more delocalized quinoid (bipolar) structure, driven by solvothermal treatment.[33] The transformation

suggests an increased conjugation length and enhanced charge delocalization, both of which contribute to higher carrier mobility and thus significantly improved conductivity. The red shift follows the order MeOH-80 > DMF-100 > DMSO-100 due to the smaller molecule MeOH more readily penetrating into the bulk of the thick film, thereby inducing the quinoid formation. The trend is consistent with the observed conductivity.

**Figure 3b** displays the XPS data of PEDOT:PSS thick films subjected to solvothermal treatment. The residual PSS ratios, calculated from the integrated area of the PSS peaks, are 33.59% (MeOH-80), 29.33% (DMF-100), and 25.21% (DMSO-100). Namely, the PSS removal rates are 66.41% (MeOH-80), 70.67% (DMF-100), and 74.79% (DMSO-100), respectively. This is because DMSO and DMF exhibit higher polarity, which enhances their molecular penetration into PEDOT:PSS thick films and facilitates the efficient removal of insulating PSS. These results confirm that solvothermal treatment promotes substantial PSS removal. PSS serves as an insulating matrix and its removal consequently significantly enhances the conductivity of the film.[34] Meanwhile, the removal of insulating PSS exposes more PEDOT chains and creates nano/micro-pores within the film.[35] However, the observed conductivity trend does not strictly follow the PSS removal ratio. For instance, although DMSO-100 exhibits the highest PSS removal, its conductivity is lower than that of MeOH-80. This discrepancy indicates that conductivity is not solely governed by PSS content. The molecular orientation, crystallinity, and  $\pi$ - $\pi$  stacking of PEDOT chains also play critical roles in determining charge transport behavior.

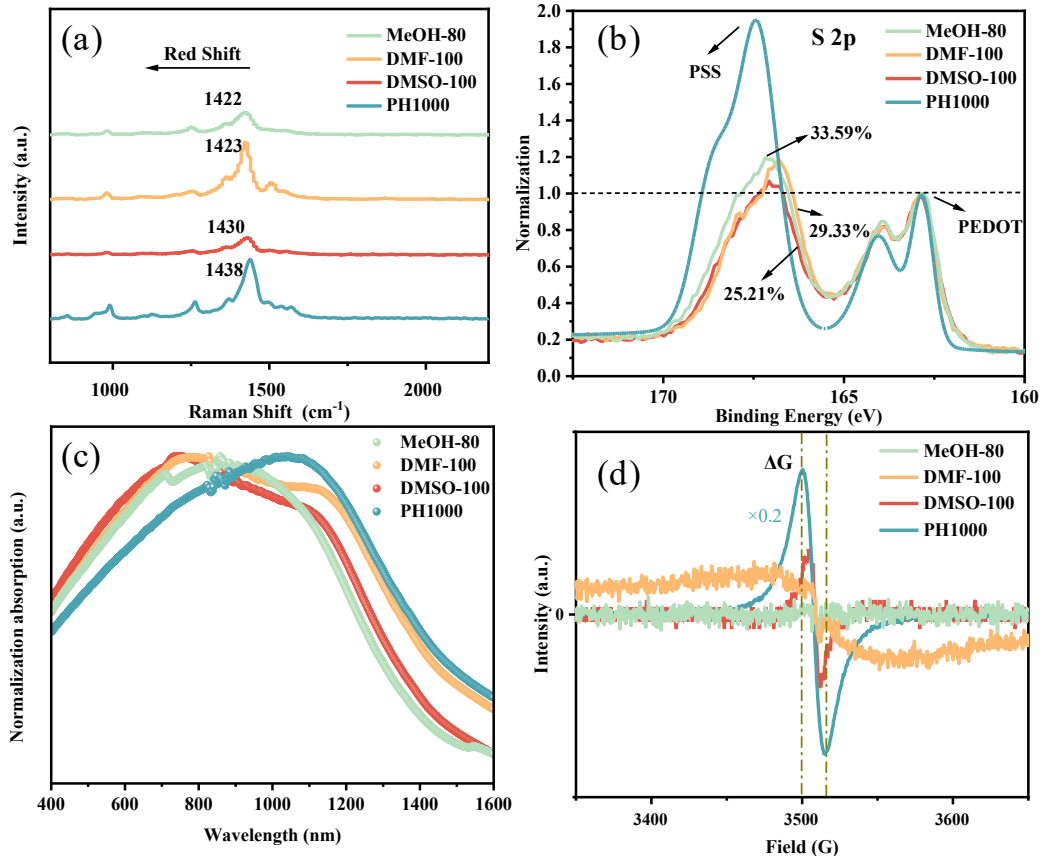


**Figure 3** (a) Raman spectra of MeOH-80, DMF-100, DMSO-100, and PH1000, and (b) XPS S 2p high-resolution spectra, (c) UV-*vis* absorption spectra and (d) ESR spectra of MeOH-80, DMF-100, DMSO-100, and PH1000.

**Figure 3c** shows the UV-*vis* spectra of PEDOT:PSS films before and after solvothermal treatment. The absorption peak at 225 nm is assigned to the phenyl group in PSS.[36] After solvothermal treatment with MeOH, DMF, and DMSO, the absorbance in this region decreased significantly, indicating a reduction in PSS content. The decreasing trend in absorbance follows the order: DMSO-100 > DMF-100 > MeOH-80, which aligns well with the XPS results, further confirming solvent-induced PSS removal. In the visible light region (400-800 nm), minimal spectral changes are observed, indicating that the PEDOT backbone remains optically stable after treatments. However, a strong and broad absorption peak appears around 1100 nm for all solvothermal-treated samples. This peak corresponds to polaron and bipolaron transitions, which are associated with the oxidized (bipolarized) state of the PEDOT chains in the near-infrared region.[37,38] The strong absorbance in near-infrared band indicates a higher concentration of charge carriers and bipolarons, directly correlating

with enhanced conductivity. This result demonstrate that, in addition to PSS removal, the bipolarons are formed during the solvothermal treatment.

**Figure 3d** presents the ESR spectra of PEDOT:PSS thick films treated with different solvents. In PEDOT:PSS, a polaron corresponds to a singly charged species accompanied by a local distortion of the polymer backbone.[39] Whereas the removal of two electrons yields a bipolaron, a spin-zero bound state with greater charge delocalization.[38] As shown in **Figure 3d**, the untreated PH1000 thick film displays a strong ESR signal, indicating that polarons dominate as charge carriers.[40] After solvothermal treatment, all films exhibit a pronounced decrease in ESR signal intensity, reflecting a transition from polaronic to bipolaronic charge carriers. This conversion is beneficial for charge transport, as bipolarons possess stronger delocalization and transportation capabilities than those of polarons. Among the treated films, MeOH-80 exhibits the lowest ESR signal, implying the highest bipolaron concentration, which is consistent with its greater conductivity. Overall, the ESR results demonstrate that the carrier form in PEDOT:PSS can be transformed from polarons to bipolarons by adjusting the processing solvents, thus leading to the conductivity enhancement of the PEDOT:PSS film in combination with the removal of PSS component and improved PEDOT stacking.

EFM measurement was employed to simultaneously map the localized conductivity distribution and surface morphology of PEDOT:PSS films (**Figure 4a-d**). In the images, bright regions correspond to conductive PEDOT phase, while dark regions indicate the insulating PSS matrix. For the untreated PH1000 film, PEDOT and PSS are uniformly dispersed. After solvothermal treatment, however, PEDOT phase becomes assembled into more defined and concentrated conductive domains-a key structural feature that contributes to the enhanced conductivity. The nanoscale conductive topographies differ with solvent type: MeOH-80 produces a dot-like distribution, DMF-100 exhibits a linear network, and DMSO-100 displays block-like conductive domains. The distinct morphology is likely synergistically governed by the Hansen solubility parameters matching and boiling point of the solvent (**Table S1**). MeOH exhibits poor PSS compatibility and low boiling point, which restricts phase

separation and produces a compact dot-like structure with high conductivity but insufficient ion channels and low capacitance. DMF achieves moderate solubility matching and constructs linear conductive networks, balancing electronic conduction and ion diffusion. With superior PSS compatibility and high boiling point, DMSO induces sufficient phase separation to form block-like porous architectures, thereby effectively improving the capacitive performance. The aggregation and alignment of PEDOT phase in solvothermal-treated films facilitate more efficient charge transport, thereby improving conductivity. The current roughness ( $R_q$ ) of PH1000, MeOH-80, DMF-100, and DMSO-100 is 247.24, 719.54, 338.97, and 976.77 fA, respectively, confirming that all solvothermal treatments enhance conductivity compared to the untreated film.

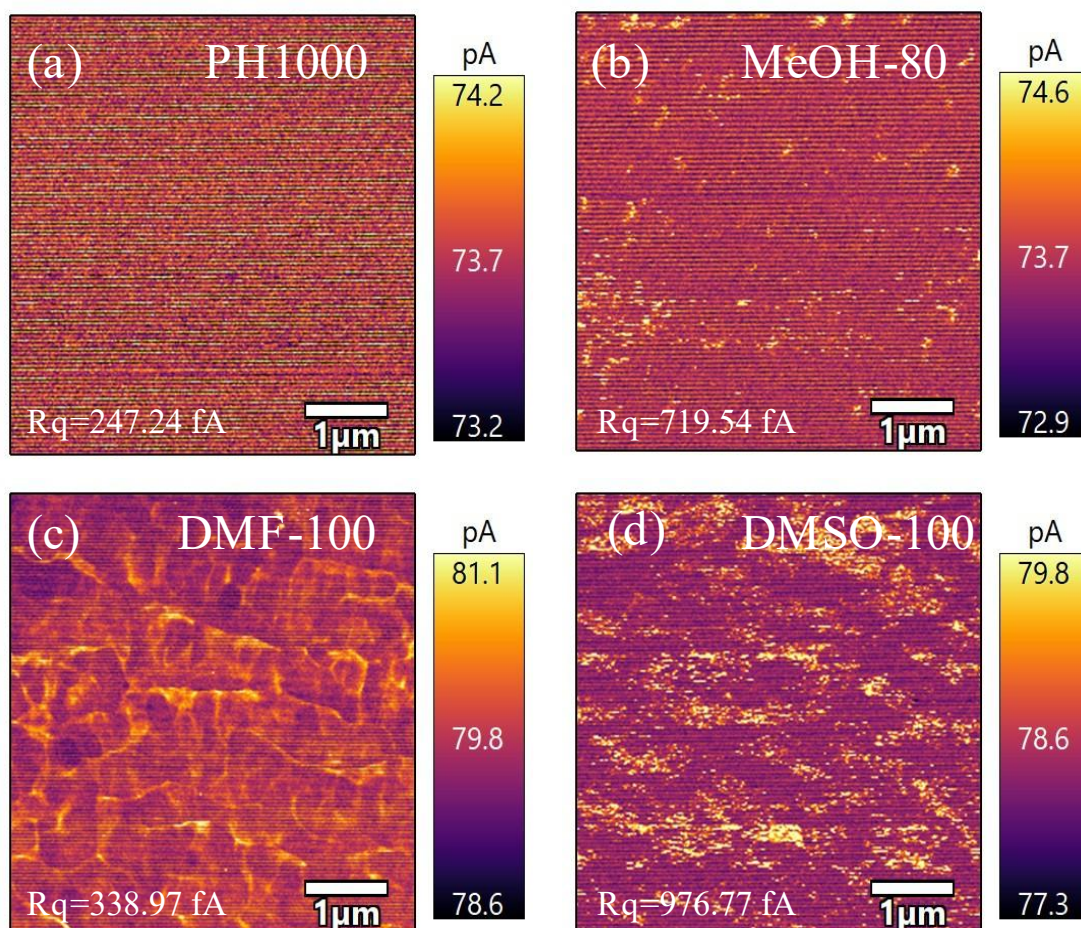


**Figure 4** EFM current mapping images of (a) PH1000, (b) MeOH-80, (c) DMF-100, and (d) DMSO-100.

The remarkable conductivity enhancement (up to  $2958 \text{ S cm}^{-1}$ ) arises from a synergistic interplay of multiscale structural reorganizations. First, the solvothermal-derived self-generated pressure enables deep penetration of polar solvents (MeOH/DMF/DMSO) into thick films, leading to effective removal of insulating PSS (25-34% reduction, XPS) and the aggregation of PEDOT phase into interconnected percolation pathways (EFM). This process minimizes charge-hopping barriers and establishes long-range conductive networks. Second, at the molecular level, the simultaneous high-temperature/pressure environment facilitates (i) enhanced crystallinity and tighter  $\pi$ - $\pi$  stacking (evidenced by reduced d-spacing along (010), GIWAXS), which act as highway channels for charge transport; (ii) a conformational transition from the benzene to the quinoid structure (Raman red shift) accompanied by a carrier form transition from polarons to highly delocalized bipolarons (quenched ESR signal). Together, these changes boost intrinsic carrier mobility along the PEDOT backbone.

Notably, the superior conductivity of MeOH-80 ( $2958 \text{ S cm}^{-1}$ ), despite its lowest PSS removal (66.41%), highlights the dominant role of microstructure optimization. Specifically, its highest crystallinity, tightest  $\pi$ - $\pi$  stacking, and higher bipolaron concentration, play a more dominant role than mere PSS extraction. This work establishes solvothermal engineering as a potent tool for decoupling and optimizing these critical parameters-PSS removal, microstructural order, and electronic carrier type in conductive polymers, thereby offering a versatile route to high-performance organic electronic materials.

## 3.2 Capacitance

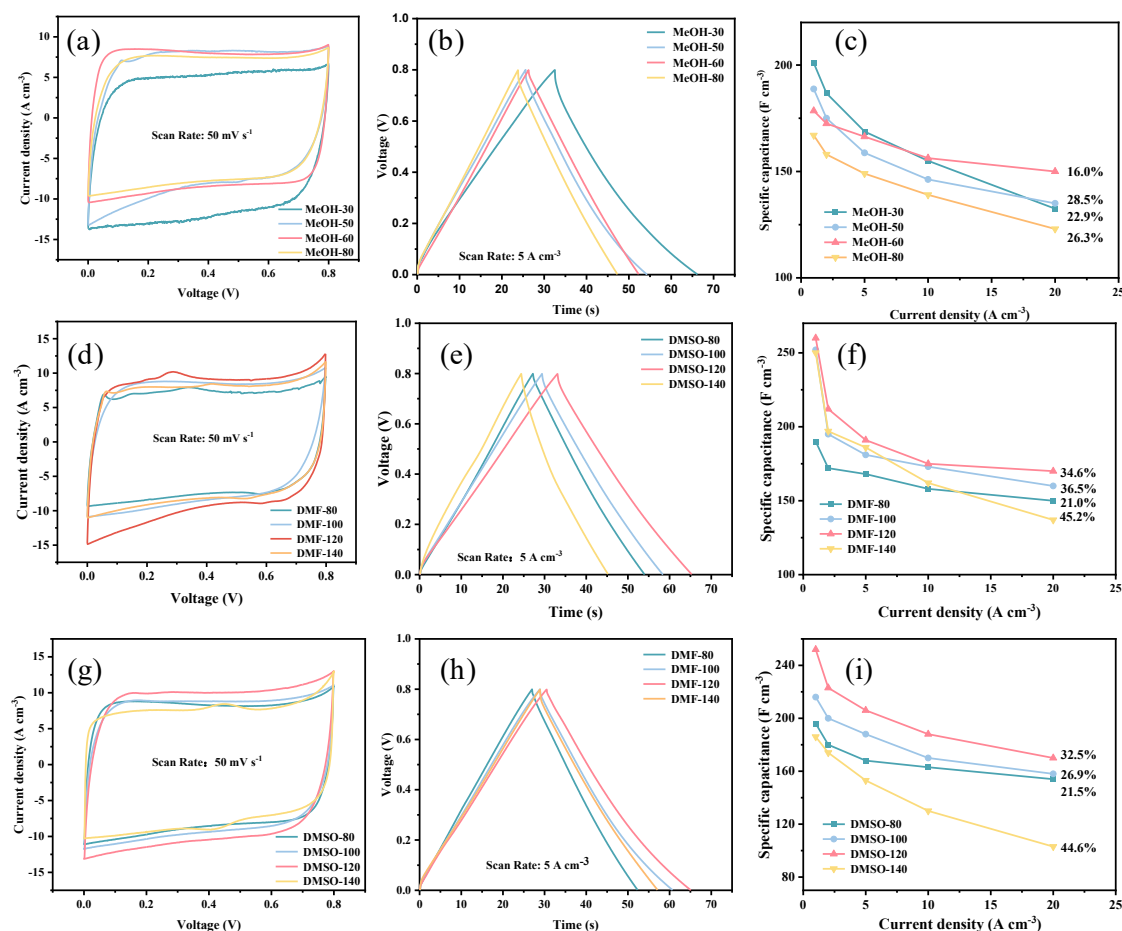


**Figure 5** CV, GCD, and specific capacitances of (a-c) MeOH-treated samples, (d-f) DMF-treated samples, and (g-i) DMSO-treated samples.

In addition to conductivity, the electrochemical properties of solvothermal-treated PEDOT:PSS films were evaluated (**Figure 5**). The CV curves of all treated samples exhibit a nearly rectangular shape, indicating their excellent capacitance response. GCD measurements were carried out at a current density of  $5 \text{ A cm}^{-3}$  (**Figure 5b, e, h**). The curves are highly symmetrical, indicative of a stable and reversible charge storage process. The rate capability was further assessed by comparing capacitance retention at a high current density of  $20 \text{ A cm}^{-3}$ . The minimum capacitance loss rates were only 16.0% (MeOH-80), 21.0% (DMF-100), and 21.5% (DMSO-100), exhibiting a great rate capability. Specifically, MeOH-treated samples are only less than 30% (**Figure 5c, f, i**). For instance, the maximum capacitance could reach  $251 \text{ F cm}^{-3}$  for DMSO-100. Even under a high current density of  $20 \text{ A cm}^{-3}$ , the capacitances of almost all samples are

still above  $130 \text{ F cm}^{-3}$ , which is significantly higher than that of the untreated PEDOT:PSS thick film.

Given that MeOH-80, DMF-100, and DMSO-100 exhibited high electrical conductivity, their electrochemical capacitances were further investigated in detail (**Figure 6a-f**). CV measurements recorded at scan rates of 10-100  $\text{mV s}^{-1}$  display nearly rectangular shapes, confirming the excellent capacitive behavior. GCD measured over a current density range of 1-20  $\text{A cm}^{-3}$  remains highly symmetrical, indicating stable and reversible charge storage. The specific volumetric capacitances, calculated from the GCD curves at  $1 \text{ A cm}^{-3}$ , were  $180 \text{ F cm}^{-3}$  for MeOH-80,  $250 \text{ F cm}^{-3}$  for DMF-100, and  $251 \text{ F cm}^{-3}$  for DMSO-100. DMF and DMSO have larger molecules with higher polarity and exhibit strong affinity for PSS, driving significant phase separation and segregation of PSS during solvothermal process (verified by XPS data). This creates a higher roughness morphology (**Figure S3**) and a three-dimensional interconnected channel network. The structure enables the electrolyte to fully infiltrate the entire electrode and significantly enhances the storage capacity. These values represent significant improvements compared to the untreated films and are also notably higher than those achieved by traditional treatment methods. For example, the DMSO-100 sample outperforms previously reported DMSO-treated PEDOT:PSS films, as shown in **Figure S4**.

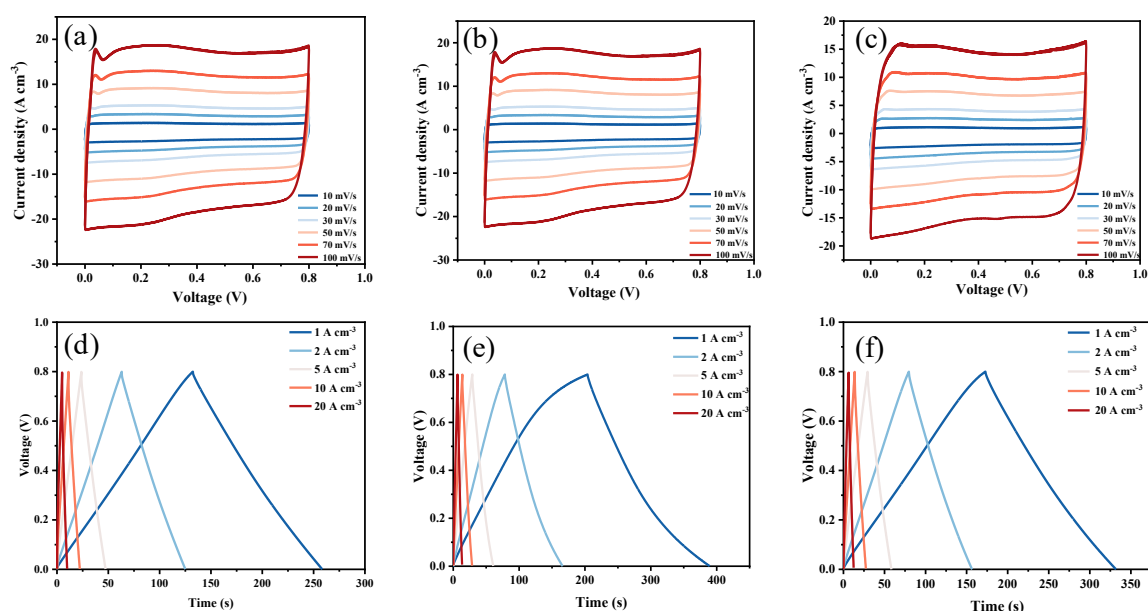


**Figure 6** (a-c) CV curves of MeOH-80, DMF-100 and DMSO-100 films at different scan rates; (d-f) GCD curves of MeOH-80, DMF-100 and DMSO-100 films at different current densities.

This remarkable capacitance performance stems from solvothermal-induced structural reorganization. Substantial removal of insulating PSS (25-34%, from XPS data) exposes more PEDOT chains and creates nano/micro-pores within the film. And EFM measurements reveals distinct conductive domain morphologies (**Figure 3**). The structure supports the interconnected internal channels and pore volume accessible to electrolyte ions. Meanwhile, solvothermal treatment fundamentally modifies the redox activity of PEDOT chains. Raman spectroscopy confirms the transition from benzoid (localized) to quinoid (delocalized) structures (**Figure 4a**), while UV-vis and ESR spectra verify the formation of highly conductive bipolaronic states (**Figure 4c,d**). The quinoid conformation, being more electrochemically active, facilitates deeper and faster pseudocapacitance behavior. Together, these multiscale structural and electronic optimizations enable the solvothermal-treated PEDOT:PSS thick films to exhibit an (a) significantly enhanced capacitance compared with pristine PEDOT:PSS.

Specifically, DMSO-100, with the highest PSS removal (74.79%), creates abundant pores/ion channels and its “block-like” conductive domains offer a favorable balance between electronic connectivity (good conductivity of  $2438 \text{ S cm}^{-1}$ ) and large, accessible interfacial area for ion exchange. Meanwhile, DMF, with a 70.67% insulating PSS removal, forms a linear network morphology that facilitates efficient charge transport while maximizing the double-layer capacitance (EDLC) contribution. In contrast, MeOH-80, characterized by the "dot-like" morphology (**Figure 3b**) and a lower PSS removal (66.41%) offers the highest conductivity ( $2958 \text{ S cm}^{-1}$ ) but provides more limited ion penetration and interfacial contact area. Consequently, the capacitance trend of solvothermal-treated PEDOT:PSS thick films is  $\text{DMSO-100} \approx \text{DMF-100} > \text{MeOH-80}$ . These results underscore that while high conductivity is essential for rapid charge/discharge and excellent rate capability, maximizing capacitance depends on optimizing the electrode-electrolyte interface through controlled PSS removal, favorable morphology, and efficient ion diffusion pathways.

**Table 2** summarizes the main performance of untreated and solvothermal-treated PEDOT:PSS films. Compared to the PH1000 films, the conductivity increases from 2 to 2958 S cm<sup>-1</sup>, and the capacitance increases from ~100 to 251 F cm<sup>-3</sup>. The solvothermal treatment not only enhances conductivity but also enhances the capacitance.

**Table 2** The resistivity, thickness, and conductivity of PEDOT:PSS films treated with different solvents.

|                                    | PH1000     | MeOH-80     | DMF-100     | DMSO-100    |
|------------------------------------|------------|-------------|-------------|-------------|
| BP (°C)                            | /          | 64.7        | 152.8       | 189         |
| Dielectric Constant                | /          | 32.7        | 36.7        | 47          |
| Resistance (Ω/□)                   | 1250± 0.02 | 1.02 ± 0.09 | 1.60 ± 0.05 | 1.27 ± 0.11 |
| Thickness (μm)                     | 4.00±0.01  | 3.64 ± 0.06 | 3.71 ± 0.05 | 3.52 ± 0.02 |
| Conductivity (S cm <sup>-1</sup> ) | 2          | 2958        | 2528        | 2438        |
| Capacitance (F cm <sup>-3</sup> )  | ~100       | 180         | 251         | 250         |

## Conclusions

In summary, this work establishes solvothermal engineering as a universal and powerful strategy for synergistically enhancing both the electrical and electrochemical properties of free-standing PEDOT:PSS thick films. By leveraging the unique high-temperature and self-generated pressure environment within a sealed system, this approach overcomes the intrinsic limitation of conventional solvent treatments in penetrating thick films, enabling deep structural reorganization. Using three common solvents (MeOH, DMF, and DMSO), we demonstrate that solvothermal treatment simultaneously optimizes multiple critical factors: substantial removal of insulating PSS, conformational transition of PEDOT chains from benzoid to quinoid structures, enhanced crystallinity and  $\pi$ - $\pi$  stacking, aggregation of PEDOT nanocrystals, and a shift toward high-mobility bipolaronic charge carriers. These synergistic effects

collectively yield record-high conductivity (up to  $2958 \text{ S cm}^{-1}$ ) and exceptional volumetric capacitances (up to  $251 \text{ F cm}^{-3}$ ), far surpassing most reported values for solvent-treated PEDOT:PSS thick films. These findings provide a foundational framework for designing high-performance conductive polymer electrodes.

## **Authorship contributions**

Jiawei Wu and Wanying Zhang wrote the main manuscript text and contributed to conceptualization. Zhen Su acquired funding, provided resources, performed writing-review & editing, and together with Tao Chen, Yingzhi Jin and Jiaxing Song developed the software. Wentao Wang performed validation. Jiayi Zeng conducted investigation. Lin Hu and Xinxing Yin provided resources. Erjun Zhou and Zaifang Li supervised the study; Zaifang Li also provided resources, performed writing-review & editing and contributed to conceptualization. All authors reviewed the manuscript.

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## **Data availability**

Data will be made available on request.

## Competing Interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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