
Research article

Sulfonated sodium alginate-doped PEDOT conductive antifreeze double-network hydrogel for flexible strain sensing

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Abstract: Conductive hydrogels, which integrate high mechanical flexibility with electrical conductivity, have attracted considerable attention for applications in flexible electronic skins and wearable health-monitoring systems. Poly(3,4-ethylenedioxythiophene) (PEDOT) is widely utilized in conductive hydrogels and flexible electronic devices owing to its excellent electrical conductivity, chemical stability, and environmental durability. However, the poor dispersibility, severe aggregation tendency, and limited film-forming capability of conventional PEDOT, together with the freezing-induced brittleness of conventional hydrogels at low temperatures, significantly restrict their practical applications. Herein, PEDOT was doped and stabilized using sulfonated sodium alginate (SSA), and poly(vinyl alcohol) (PVA), polyacrylamide (PAM), ethylene glycol (EG), and an ionic liquid (IL) were introduced to construct a PVA/PAM/EG/PEDOT:SSA/IL (PPEPSI) conductive double-network hydrogel with excellent freeze resistance. The results demonstrated that SSA effectively inhibited

PEDOT aggregation. When the 3,4-ethylenedioxythiophene (EDOT)-to-SSA feed ratio was optimized to $1.0 \text{ mL} \cdot \text{g}^{-1}$, the average particle size of the PEDOT dispersion was reduced to $0.32 \text{ } \mu\text{m}$, accompanied by an electrical conductivity of $8.12 \text{ S} \cdot \text{m}^{-1}$. The resulting PPEPSI hydrogel exhibited a dense three-dimensional network structure. With the incorporation of 10 wt% IL, the hydrogel achieved a tensile strength of 436 kPa, an elongation at break of 218%, and an electrical conductivity of 21.65 mS/cm . Furthermore, owing to the synergistic effects of EG and IL, the hydrogel maintained excellent flexibility at $-20 \text{ }^{\circ}\text{C}$. As a flexible strain sensor, the hydrogel exhibited a maximum gauge factor (GF) of 3.06 over a strain range of 0–240%, with response and recovery times of 0.216 and 0.243 s, respectively. The sensor enabled real-time monitoring of multiscale human motions and handwriting signals, while structural reconstruction of the hydrogel network was achieved through thermal and low-temperature treatments. In addition, the hydrogel precursor exhibited shear-thinning behavior, enabling the fabrication of complex patterns via 3D printing. These results demonstrate the potential of the PPEPSI hydrogel for applications in flexible electronics and smart wearable devices.

Keywords: strain sensor; conductive hydrogel; PEDOT; freeze resistance; human motion monitoring

1. Introduction

With the rapid development of flexible electronics, wearable devices, electronic skin, and intelligent human–machine interaction technologies, flexible sensing materials capable of delivering stable signals under complex deformations have attracted increasing attention [1–4]. Conductive hydrogels combine high water content, excellent flexibility, biocompatibility, and tunable electrical conductivity, while exhibiting mechanical properties comparable to those of soft biological tissues. These characteristics make them promising materials for flexible sensors, health-monitoring devices, and smart wearable systems [5–7], as well as for three-dimensional biofabrication platforms requiring dynamically tunable mechanics [8]. Accordingly, conductive hydrogel-based strain sensors have been extensively investigated for human-motion monitoring, physiological-signal detection, electronic skin, and soft robotics [9–11]. Nevertheless, conventional conductive hydrogels still suffer from unstable conductive networks, inadequate mechanical robustness, and freezing-induced performance degradation at low temperatures, which severely limit their practical applications in complex environments [12–15]. Multifunctional hydrogels with rigid–flexible synergy [16], poly(3,4-ethylenedioxythiophene) (PEDOT)-based foam–hydrogel triboelectric systems [17], and all-hydrogel yarn-based sensors [18] have advanced wearable sensing and energy harvesting, yet simultaneously achieving high conductivity, mechanical robustness, and freeze resistance remains challenging.

Currently, the conductive mechanisms of conductive hydrogels mainly involve two modes: electronic conduction and ionic conduction. Common conductive components include carbon nanotubes, graphene, MXene, and conductive polymers [19–22]. Among them, PEDOT has been widely utilized in conductive hydrogels and flexible electronic systems owing to its high electrical conductivity, excellent chemical stability, and environmental durability [23–26]. However, strong π – π stacking and hydrophobic interactions between PEDOT chains promote aggregation in aqueous environments and hydrophilic polymer matrices. This aggregation hinders the formation of uniform and continuous conductive networks, thereby deteriorating the electrical properties, mechanical

performance, and sensing stability of composite hydrogels [27–30]. Therefore, developing effective PEDOT doping and dispersion strategies is crucial for the fabrication of high-performance conductive hydrogels [31–34]. Commercial poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) systems have demonstrated impressive performance in demanding scenarios such as high-resolution electromyography (EMG) monitoring [35], implantable bioelectronics [36], and underwater robotic sensing [37,38].

Currently, PSS is one of the most commonly used dopants and dispersing agents for PEDOT. PEDOT:PSS systems have been extensively applied in flexible electronics and conductive hydrogels owing to their excellent electrical conductivity and processability [39–42]. However, PSS is a petroleum-derived polymer, and its environmental sustainability and biocompatibility remain relatively limited. In contrast, natural polymers have attracted considerable attention because of their abundance, renewability, excellent biocompatibility, and structural tunability [43–46]. Sodium alginate (SA) is a typical natural polysaccharide with excellent hydrophilicity, biocompatibility, and gel-forming ability, and it has been widely used in flexible sensing, biomedical materials, and tissue engineering [43,44]. However, SA intrinsically lacks sufficient functional groups for strong interactions with PEDOT. Sulfonation modification introduces sulfonic acid groups into the SA molecular chain, resulting in sulfonated sodium alginate (SSA). SSA can interact with PEDOT segments through electrostatic interactions, while its hydrophilic polymer backbone provides steric hindrance, thereby effectively suppressing PEDOT aggregation, improving dispersion stability in aqueous solutions, and enhancing the uniformity of conductive networks. Previous studies have demonstrated that biomacromolecules and natural polymers containing sulfonic acid groups can serve as environmentally friendly doping or dispersing agents for PEDOT, effectively improving its electrical conductivity and dispersion stability [31,32,47,48]. Therefore, SSA represents a promising green and highly efficient biomass-derived dopant for PEDOT.

In addition to electrical conductivity, stable operation under low-temperature conditions is another critical requirement for practical flexible sensors. Conventional hydrogels contain large amounts of free water, which can easily form ice crystals at low temperatures, resulting in network damage, reduced flexibility, and even loss of electrical conductivity [12,49,50]. Recently, the incorporation of organic solvents, salts, or ionic liquids into hydrogel networks has emerged as an effective strategy for improving freeze resistance and low-temperature adaptability [51–54]. Ionic hydrogels integrating high conductivity, self-healing, and adhesion have further demonstrated reliable health monitoring and human–machine interactions under extreme cold conditions [55]. Among these strategies, ethylene glycol (EG) can reduce the freezing point of hydrogel systems by forming hydrogen bonds with water molecules and improving moisture retention [50]. Ionic liquids, meanwhile, exhibit low volatility, high ionic conductivity, and excellent low-temperature stability. They not only suppress ice crystal formation but also regulate the conformation of conductive polymer segments through enhanced ion transport and secondary doping effects, thereby improving the synergistic transport of electrons and ions [52–54,56,57]. Therefore, integrating ionic liquids with conductive polymers to construct conductive antifreeze hydrogels is expected to simultaneously enhance mechanical properties, electrical performance, and low-temperature stability. While PEDOT-based hydrogels have advanced toward fatigue-resistant underwater sensing [37], wireless electrotactile interfaces [58], and wearable bioelectronics [59,60], most systems still rely on petroleum-derived PSS and fail to simultaneously achieve high conductivity, mechanical robustness, and freeze resistance.

In this study, SSA was employed as a dopant and dispersing agent for PEDOT to prepare PEDOT:SSA conductive composites, which were subsequently incorporated into a poly(vinyl alcohol) (PVA)/polyacrylamide (PAM) double-network system. By introducing EG and the ionic liquid [BMIM][DCA], a PVA/PAM/EG/PEDOT:SSA/ionic liquid (IL) (PPEPSI) conductive antifreeze hydrogel was constructed. The structure and electrical conductivity of PEDOT:SSA composites were systematically investigated, together with the effects of ionic liquid content on the hydrogel microstructure, mechanical properties, electrical properties, freeze resistance, and sensing performance. Furthermore, the application potential of the hydrogel in human motion monitoring, handwriting identification, and 3D printing was demonstrated.

2. Materials and methods

2.1. Materials

Chlorosulfonic acid (CSA, CP), acetone (CP), and N,N-dimethylformamide (DMF, CP) were purchased from Shaanxi Dexiang Laboratory Equipment Co., Ltd. PVA ($\geq 99.0\%$) was purchased from Sinopharm Chemical Reagents Co., Ltd. SA (AR), EDOT (99%), acrylamide (AM, AR, 99%), 1-butyl-3-methylimidazolium dicyanamide ([BMIM][DCA], 98%), ammonium persulfate (APS, AR, 98.5%), and N,N'-methylenebisacrylamide (MBA, AR) were purchased from Shanghai Macklin Biochemical Co., Ltd. Ethylene glycol (EG, AR) was purchased from Tianjin Tianli Chemical Reagents Co., Ltd. All chemicals were used as received without further purification, and deionized water was used throughout all experiments.

2.2. Preparation of PEDOT:SSA

First, SA was sulfonated using CSA to prepare SSA. Specifically, DMF was slowly added to CSA, and the mixture was reacted in an ice-salt bath. Subsequently, SA was added, and the reaction was continued at 60 °C for 4 h. The resulting product was precipitated with acetone, dialyzed, and freeze-dried to obtain SSA powder. Subsequently, PEDOT:SSA composites were prepared via in situ oxidative polymerization using SSA as a dopant and dispersing agent. The obtained SSA powder was dissolved in deionized water, and different amounts of EDOT monomer (0.5, 0.75, 1.0, and 1.25 mL) were added. After ultrasonic dispersion, APS was introduced as an oxidizing agent, and the mixture was polymerized at room temperature for 48 h. The resulting product was precipitated, dialyzed, and freeze-dried to obtain PEDOT:SSA powder. A control PEDOT sample was prepared using the same procedure without the addition of SSA.

2.3. Preparation of PPEPSI conductive double-network freeze-resistant hydrogel

PVA was dissolved in deionized water and heated under stirring at 90 °C until a homogeneous and transparent solution was obtained. The PVA solution was then mixed with the EG/PEDOT:SSA dispersion, followed by the addition of different concentrations of the ionic liquid [BMIM][DCA] (0%, 5%, 7.5%, 10%, and 12.5%). After sufficient stirring, AM, MBA, APS, and tetramethylethylenediamine (TMEDA) were sequentially added, and the precursor solution was rapidly injected into the mold. Freeze-thaw cycles were performed by freezing at -24 °C for 12 h and thawing at room temperature

for 4 h. A total of five cycles were conducted to obtain the PPEPSI conductive double-network freeze-resistant hydrogel. For clarity, the hydrogels are denoted as follows: PPE (PVA/PAM/EG, without PEDOT:SSA or IL), PPEPS (PVA/PAM/EG/PEDOT:SSA, with PEDOT:SSA but without IL), and PPEPSI (PVA/PAM/EG/PEDOT:SSA/IL, the complete formulation).

2.4. Preparation of PPEPSI hydrogel sensors

The prepared PPEPSI hydrogels were cut into strip-shaped specimens, and copper electrodes were attached to both ends to fabricate flexible strain sensors. During testing, the sensors were connected to a digital source meter, and their strain-sensing performance was evaluated by recording real-time resistance changes. In human motion monitoring experiments, the sensors were attached to the face, fingers, wrists, and elbows. The relative resistance variations under different motion states were collected to evaluate their human motion sensing capability.

2.5. Characterization and performance testing

A scanning electron microscope (SEM, SU-8010) was used to observe the morphologies of PEDOT, PEDOT:SSA, and freeze-dried PPEPSI hydrogels. An energy-dispersive X-ray spectrometer (EDS) was employed to analyze the elemental distribution of the PPEPSI hydrogel. Fourier-transform infrared spectroscopy (FTIR, 4000–400 cm^{-1}) and X-ray photoelectron spectroscopy (XPS, Thermo Scientific K-Alpha, Al-K α) were used to characterize the functional groups, elemental compositions, and chemical states of PEDOT, PEDOT:SSA, and PPEPSI hydrogels. The thermal stability of SSA, PEDOT, PEDOT:SSA, and PPEPSI hydrogels was analyzed using a thermogravimetric analyzer (TGA) under a nitrogen atmosphere. The samples were heated from room temperature to 700 $^{\circ}\text{C}$ at a heating rate of 10 $^{\circ}\text{C}\cdot\text{min}^{-1}$. The electrical conductivity of PEDOT:SSA films was measured using an RTS-9 four-point probe system, and the particle size distributions of PEDOT and PEDOT:SSA dispersions were determined using a laser particle size analyzer (LT2200). The mechanical properties of PPEPSI hydrogels were tested using a universal testing machine (Instron 5500). The specimens were prepared with dimensions of 80 mm $\times$ 20 mm and tested at a tensile strain rate of 2 $\text{mm}\cdot\text{s}^{-1}$. The phase transition behavior and freeze resistance of PPE, PPEPS, and PPEPSI hydrogels were evaluated using differential scanning calorimetry (DSC, TA) under a nitrogen atmosphere. The samples were cooled from 25 to -80 $^{\circ}\text{C}$ at a cooling rate of 5 $^{\circ}\text{C}\cdot\text{min}^{-1}$. Electrical conductivity was evaluated using electrochemical impedance spectroscopy (EIS) with a CHI 660E electrochemical workstation and a three-electrode system consisting of a platinum auxiliary electrode, saturated calomel reference electrode, and hydrogel working electrode. Measurements were performed in 1 $\text{mol}\cdot\text{L}^{-1}$ H_2SO_4 electrolyte at a test potential of 0.2 V, a frequency range of 0.1– 10^5 Hz, and a sine wave amplitude of 5 mV. Prior to testing, the hydrogels were cut into specimens with dimensions of 3 mm $\times$ 3 mm and a thickness of 1.5 mm. The conductivity was calculated according to Eq 1:

$$\sigma = \frac{d}{R \times A} \quad (1)$$

In Eq 1, σ is the conductivity, R is the resistance, d is the sample thickness, and A is the sample area; the unit of σ is $\text{mS}\cdot\text{cm}^{-1}$, the unit of d is cm, the unit of R is Ω , and the unit of A is cm^2 .

The hydrogel samples were stored at 20 ± 2 °C and $25\% \pm 3\%$ relative humidity (RH) for 20 days. The moisture retention ratio was calculated using the W_t/W_0 ratio, where W_0 represents the initial weight and W_t represents the current weight. By coupling a digital source meter (Keithley 2461) with a universal testing machine (Instron 5500), real-time resistance signals of the hydrogel were acquired under tensile and compressive deformation conditions, and resistance variations during mechanical loading were simultaneously recorded.

The real-time resistance signals of hydrogel samples were collected using a Keithley source-meter at room temperature (25 °C). During the test, both ends of the hydrogel were clamped and fixed on the test fixture. After the baseline resistance became stable, a predetermined tensile strain was rapidly applied to the sample and kept constant until the resistance signal reached a steady-state value. Subsequently, the tensile strain was quickly removed once the resistance value stabilized, allowing the sample to rebound naturally to its initial state. The dynamic evolution of resistance was recorded throughout the whole process. The sampling frequency of the source-meter was set to 50 Hz (corresponding to a temporal resolution of 20 ms).

Response time is the time required for the relative resistance change $(R-R_0)/R_0$ to rise from the baseline value to 90% of the steady-state response value, starting from the instant when tensile strain is applied. Recovery time is the time required for the relative resistance change $(R-R_0)/R_0$ to fall from the steady-state response value to 10% above the baseline value (i.e., recovery to 90% of the initial resistance), starting from the instant when tensile strain is released. Specifically, R represents the real-time resistance under tensile strain, and R_0 denotes the initial baseline resistance without applied strain.

Elemental analysis: the carbon, hydrogen, nitrogen, and sulfur content of the alginate was measured by the elemental analysis method. The degree of sulfonation (DS), the number of sulfate groups per monomer, was calculated using Eq 2 [61]:

$$DS = \frac{198[S]}{3200-102[S]} \quad (2)$$

where $[S]$ was the sulfur content (%) of sulfated alginate obtained from the element analysis.

3. Results and discussion

3.1. Preparation and characterization of PEDOT:SSA

Figure 1a illustrates the preparation process of PEDOT:SSA. First, SA was sulfonated using CSA to obtain SSA. The reaction product contains 10.83% sulfur, indicating a DS of 1.02. This infers that, of the two hydroxyl groups each repeating unit contains, on average, 1.02 are replaced by sulfate groups. Consequently, a total conversion of 51% was achieved. The DS of SSA determines sulfonic acid group density, which directly modulates PEDOT dispersion and doping level. Moderate DS enhances electrostatic stabilization and charge delocalization, whereas excessive DS may dilute conductive pathways or perturb polymerization kinetics. An optimal DS is therefore anticipated, and its quantitative relationship with composite properties will be addressed in future studies. Subsequently, PEDOT:SSA composites were prepared via in situ oxidative polymerization using SSA as a dopant and dispersing agent. To optimize the preparation conditions of the PEDOT:SSA conductive filler, samples with different EDOT:SSA feed ratios were characterized in terms of morphology and electrical properties. As shown in Figure 1b, pristine PEDOT exhibits a distinct agglomerated

morphology with densely packed particles, indicating that strong π - π interactions between PEDOT chains promote severe aggregation in aqueous environments. After the introduction of SSA, the aggregation behavior of PEDOT is significantly suppressed (Figure 1c–f). Specifically, at an EDOT:SSA ratio of 1.25 mL:1.0 g (Figure 1c), the aggregates become looser and exhibit initial structural evolution, suggesting that SSA begins to function as a soft template and dispersing agent. PEDOT aggregation is reduced through steric hindrance; however, irregular aggregates remain, and the structural continuity remains insufficient. When the EDOT:SSA ratio reaches 1.0 mL:1.0 g (Figure 1d), the sample consists of relatively uniform nanoparticles and forms an interconnected porous structure, which is beneficial for electron transport and subsequent hydrogel fabrication. The locally magnified image (Figure 1g) reveals a rough surface, close interparticle contact, and an interconnected porous stacked structure. This structure originates from the SSA-assisted templating effect during in situ polymerization, which not only promotes the formation of conductive pathways but also provides favorable interfacial conditions for subsequent hydrogel fabrication. As shown in Figure 1e,f, with further decreasing EDOT content, although the particle size decreases, excessive SSA induces stronger steric hindrance, causing the structure to become denser and more matrix-like. This reduces the relative content of the conductive PEDOT phase, which is unfavorable for the construction of an efficient conductive network.

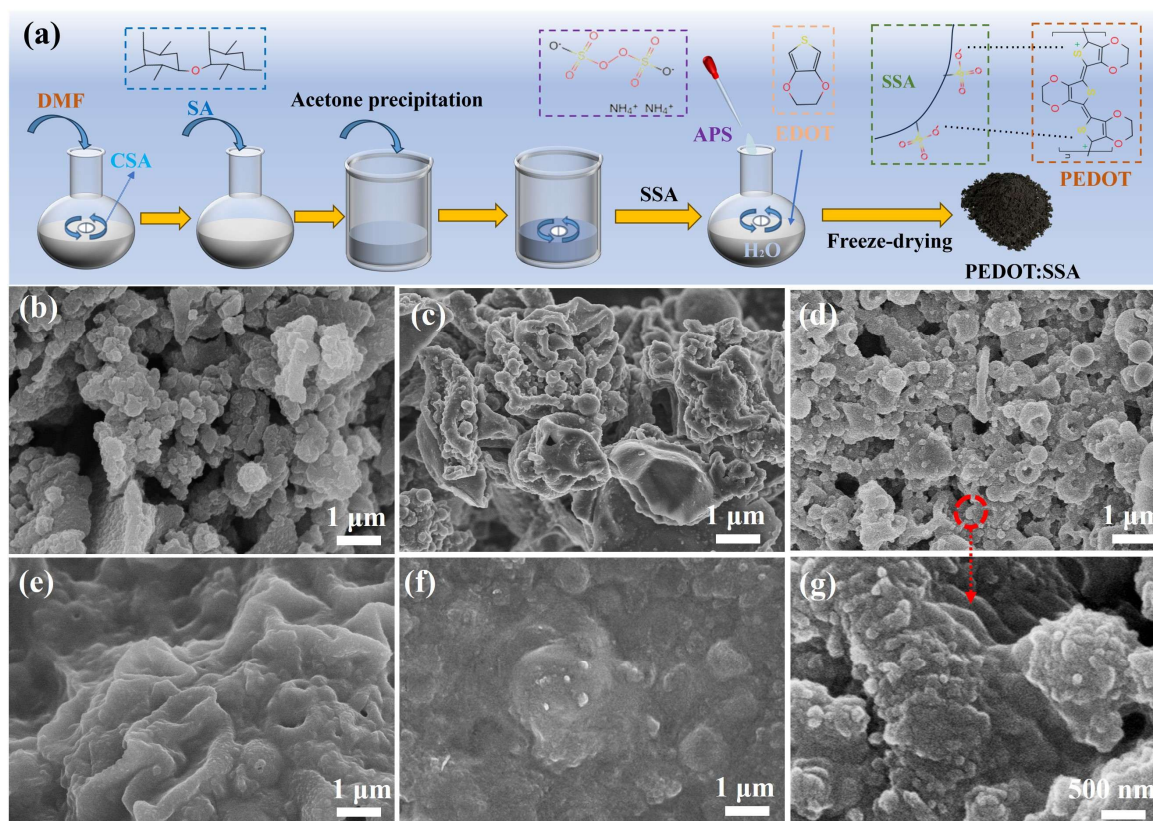


Figure 1. (a) Schematic illustration of the preparation of PEDOT:SSA. SEM images of PEDOT:SSA prepared at different EDOT:SSA ratios: (b) PEDOT; (c) EDOT:SSA = 1.25 mL:1.0 g; (d) EDOT:SSA = 1.0 mL:1.0 g; (e) EDOT:SSA = 0.75 mL:1.0 g; (f) EDOT:SSA = 0.5 mL:1.0 g. (g) Enlarged view of sample (d).

Furthermore, as shown in Figure S1, the appearance of samples prepared with different EDOT contents demonstrates that the sample color gradually changes from light brown to deep blue-black with increasing EDOT content, indicating continuous formation and growth of PEDOT chains. Dispersion stability tests (Figure S2) further demonstrate that SSA effectively improves the storage stability of the PEDOT dispersion.

The FTIR and XPS results further confirm the successful synthesis of PEDOT:SSA. As shown in Figure 2a, SSA exhibits two new characteristic absorption peaks located at approximately 1200 and 1030 cm^{-1} , which are assigned to the stretching vibrations of S=O and S–O bonds in sulfonic acid groups, respectively. These results indicate that sulfonic acid groups are successfully introduced into the sodium alginate backbone, confirming the successful synthesis of SSA. As shown in Figure 2b, compared with pristine PEDOT, the PEDOT:SSA composites prepared with different EDOT feed amounts exhibit characteristic absorption peaks near 1510 and 1358 cm^{-1} , corresponding to the stretching vibrations of C=C and C–C bonds in the thiophene rings, respectively. The absorption near 1100 cm^{-1} is attributed to the C–S vibration of PEDOT chains, while the peak at approximately 1030 cm^{-1} matches the S–O vibration region of SSA. These spectral features support the coexistence of PEDOT and SSA in the composite. Meanwhile, all samples maintain a broad hydroxyl absorption band in the range of 3200–3500 cm^{-1} , suggesting the preservation of the hydroxyl-rich SSA structure in the composite materials. The combined FTIR results in Figure 2a,b confirm that sodium alginate is successfully converted into SSA after sulfonation, and that PEDOT:SSA composites are successfully constructed through SSA-assisted oxidative polymerization of EDOT. Dynamic light scattering (DLS) results reveal that pristine PEDOT exhibits a broad and irregular particle size distribution, whereas PEDOT:SSA exhibits a narrower unimodal distribution with an average particle size of approximately 320 nm (Figure 2c). These results indicate that SSA effectively suppresses PEDOT aggregation through electrostatic interactions and steric hindrance.

Full-scan XPS spectra and high-resolution S 2p spectra (Figure 2d,e) reveal that, in addition to the thiophene sulfur signal originating from PEDOT, sulfur signals associated with the sulfonic acid groups of SSA are also observed in PEDOT:SSA. The coexistence of these two sulfur species confirms the presence of both PEDOT and SSA in the composite, which is consistent with a doped PEDOT:SSA system. The maintenance of distinct chemical states suggests that SSA acts as a counter-ion dopant rather than undergoing chemical transformation during polymerization. The thermogravimetry (TG) and derivative thermogravimetry (DTG) curves (Figure S3a,b) demonstrate that the thermal degradation behavior of PEDOT:SSA differs from those of pristine SSA and PEDOT, indicating strong interactions between the two components and enhanced thermal stability of the composite. These thermal behavior differences suggest intermolecular interactions between PEDOT and SSA, which may restrict chain mobility and alter the decomposition pathway of the composite. Figure 2f presents the electrical conductivity of PEDOT:SSA composites prepared with different EDOT feed contents. As the EDOT feed amount increases from 0.5 to 1.25 mL, the electrical conductivity increases from 0.09 to 12.29 $\text{S}\cdot\text{m}^{-1}$, indicating that the increased PEDOT content facilitates the formation of continuous conductive pathways. Meanwhile, the sulfonic acid groups in SSA facilitate charge delocalization and improve the doping efficiency of PEDOT chains, thereby enhancing charge transport. Although the sample prepared with 1.25 mL EDOT exhibits the highest conductivity, Figure S2 demonstrates that this sample possesses poor dispersion stability and is prone to aggregation and sedimentation. In contrast, the 1.0 mL EDOT sample maintains relatively high conductivity (8.12 $\text{S}\cdot\text{m}^{-1}$) while exhibiting improved dispersion stability and reproducibility. The selection of the optimal

EDOT:SSA ratio (1.0 mL:1.0 g) was based on a comprehensive evaluation of multiple factors. The 1.0 mL sample achieves high conductivity ($8.12 \text{ S} \cdot \text{m}^{-1}$, Figure 2f) and uniform nanoparticle morphology (Figure 1d), whereas the 1.25 mL sample aggregates severely within 12 days (Figure S2) despite higher conductivity, and the 0.5–0.75 mL samples are inadequately conductive ($0.09\text{--}1.16 \text{ S} \cdot \text{m}^{-1}$). Therefore, considering the balance among electrical conductivity, dispersion stability, morphological uniformity, and practical processability, the EDOT:SSA ratio of 1.0 mL:1.0 g was identified as the optimal condition for subsequent hydrogel fabrication. A concise summary of these comparative results is provided in Table S1. Compared with previously reported PEDOT:PSS, PEDOT:CS, and PEDOT:LGS systems, the PEDOT:SSA composite developed in this study exhibits higher electrical conductivity (Table 1), demonstrating that SSA effectively regulates the aggregation behavior and structural organization of PEDOT chains, thereby promoting conductive network formation.

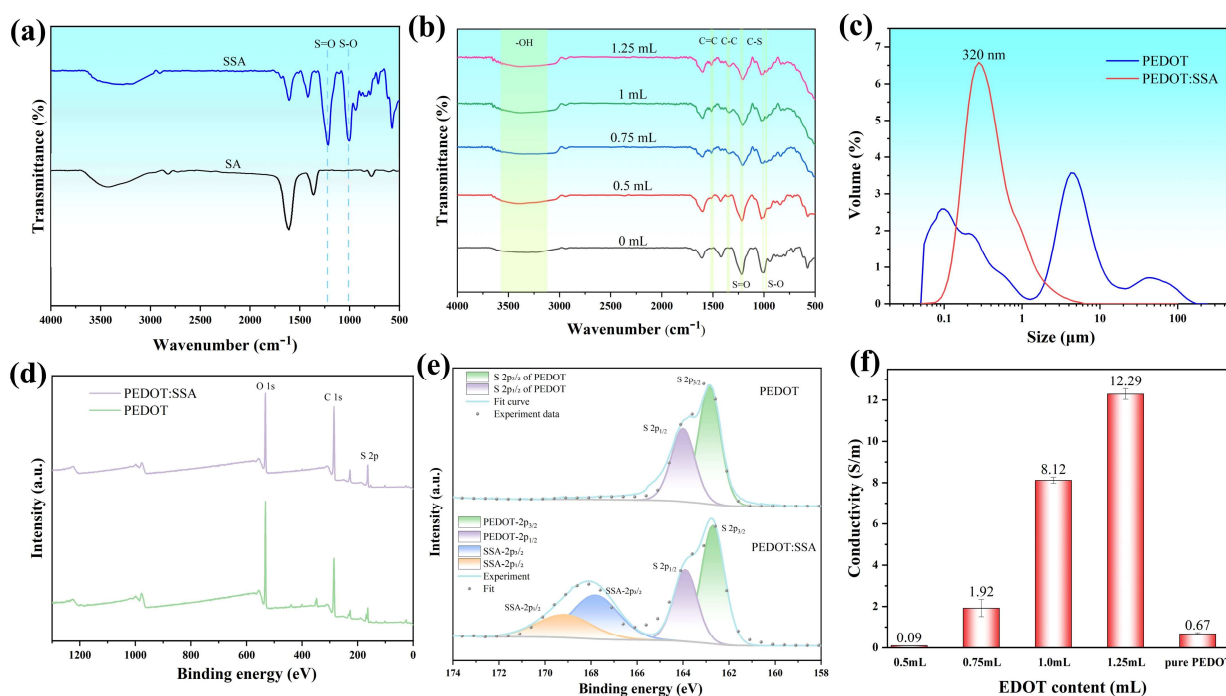


Figure 2. Structural characterization and conductivity evaluation of PEDOT:SSA. (a) FTIR spectra of SA and SSA; (b) FTIR spectra of PEDOT:SSA synthesized with different EDOT contents; (c) particle size distributions of PEDOT and PEDOT:SSA; (d) XPS survey spectra of PEDOT and PEDOT:SSA; (e) high-resolution S 2p spectra of PEDOT and PEDOT:SSA; (f) electrical conductivity of PEDOT:SSA prepared with varying EDOT contents.

In the oxidative chemical polymerization of EDOT, ammonium persulfate $(\text{NH}_4)_2\text{S}_2\text{O}_8$ serves as the initiator to trigger the progressive growth of PEDOT chains. EDOT monomers are oxidized by $\text{S}_2\text{O}_8^{2-}$ to generate cationic radicals (EDOT^+), which undergo dimerization accompanied by the elimination of two protons to form stable neutral dimers. The resulting PEDOT dimers are further oxidized by $\text{S}_2\text{O}_8^{2-}$, enabling continuous chain propagation. Persistent oxidation of PEDOT segments generates radical species with positive charge carriers (holes), necessitating the incorporation of negatively charged dopants throughout polymerization to compensate for these excess positive charges [62]. SSA, prepared by sulfonation of SA, contains numerous sulfonic acid (SO_3H) and carboxylic acid ($-\text{COOH}$)

functional groups. The SO_3H groups act as potent Brønsted acids. Upon deprotonation, they release protons (H^+) and generate negatively charged sulfonate anions (SO_3^-). The negatively charged sulfonate groups (SO_3^-) on SSA act as counter-ions to balance the positive charges (polarons and bipolarons) that form on the PEDOT backbone during oxidative polymerization [63]. As an effective macromolecular dopant, SSA establishes robust charge-trapping sites to balance the positive charges on the PEDOT backbone during polymerization, thereby preserving the favorable electrical conductivity of the PEDOT polymer network [64].

Table 1. Intrinsic electrical conductivity of PEDOT-based materials with primary doping.

Materials	Electrical conductivity	Reference
PEDOT:PSS	0.3 S/m	[31]
PEDOT:CS	0.576 S/m	[65]
PEDOT:LGS	0.0171 S/m	[66]
PEDOT:SSA	8.12 S/m	Our work

3.2. Preparation and characterization of PPEPSI hydrogels

Figure 3a illustrates the preparation process of the hydrogel. Based on the optimized PEDOT:SSA composite, it was incorporated into a PVA/PAM double-network system together with EG and [BMIM][DCA] to construct a conductive freeze-resistant PPEPSI hydrogel. SEM results demonstrate that the ionic liquid content significantly influences the pore structure of the hydrogel and the distribution of the conductive phase. As shown in Figure 3b, the hydrogel without ionic liquid exhibits relatively large pores and a loose network structure. With increasing [BMIM][DCA] content to 5% (Figure 3c), 7.5% (Figure 3d), and 10% (Figure 3e), [BMIM][DCA] acts as a dopant and plasticizer [67] effectively promoting interfacial interactions between PEDOT:SSA and the polymer matrix, facilitating uniform dispersion of the conductive phase. Consequently, the hydrogel pores gradually shrink and become denser, accompanied by reduced pore size and enhanced structural connectivity, resulting in a more uniform and stable three-dimensional network structure. Therefore, synergistic enhancement of ionic transport capability and mechanical stability is achieved. When the ionic liquid content further increases to 12.5% (Figure 3f), the local structure becomes heterogeneous, suggesting that excessive ionic liquid may interfere with the cross-linked network formation. The high-magnification image of Figure 3e reveals uniformly distributed nanoscale conductive domains on the scaffold surface (Figure 3g). This phenomenon is attributed to the enhanced doping and dispersion of PEDOT:SSA induced by the ionic liquid, which facilitates the formation of continuous conductive pathways while maintaining the integrity of the hydrogel network. EDS mapping results (Figure 3h) reveal the uniform distribution of C, N, O, and S elements throughout the hydrogel matrix, confirming the homogeneous dispersion of PEDOT:SSA within the PPEPSI hydrogel.

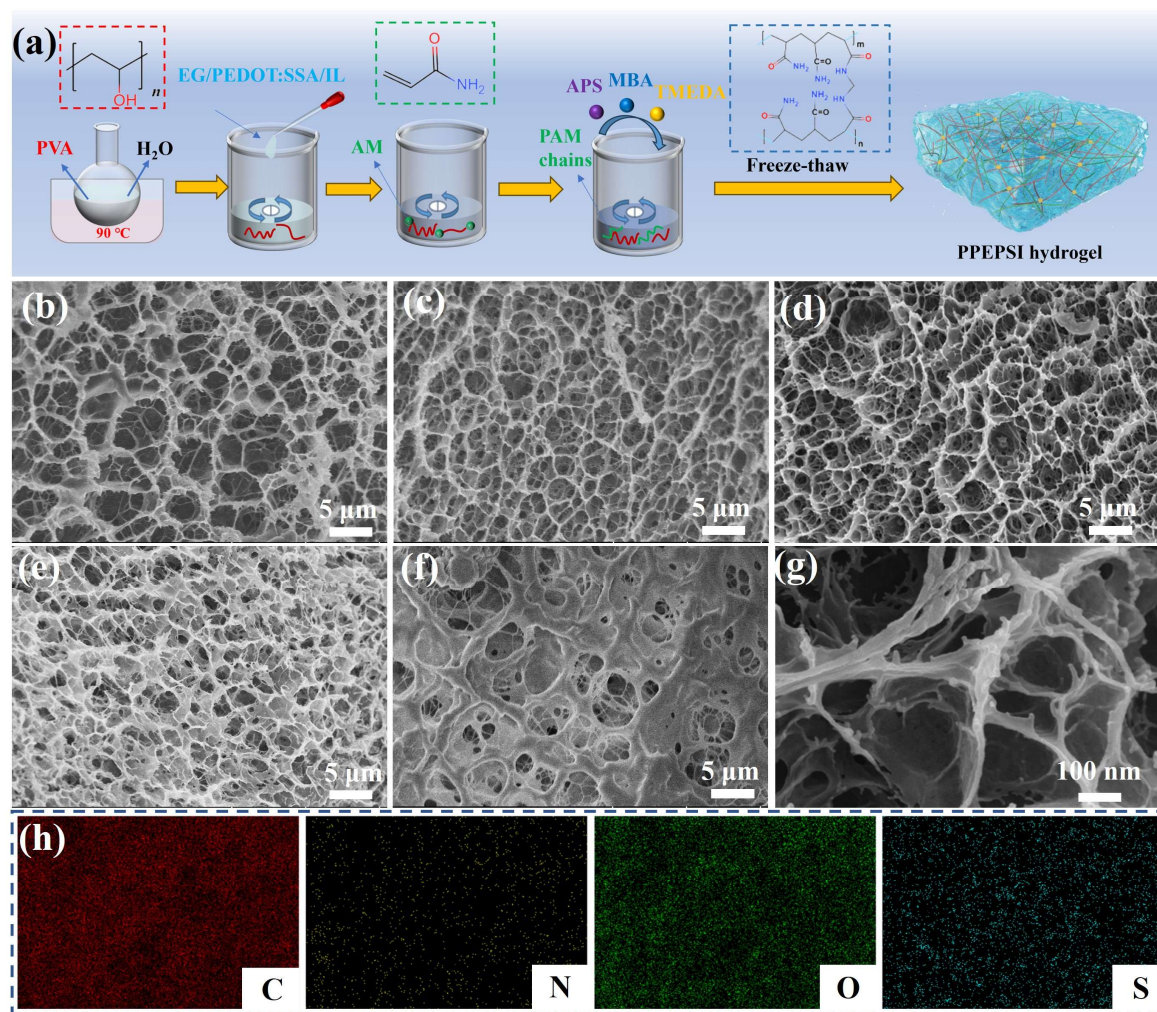


Figure 3. (a) Schematic illustration of the preparation of PEDOT:SSA. SEM images of PPEPSI hydrogels with different ionic liquid contents: (b) 0%, (c) 5%, (d) 7.5%, (e) 10%, and (f) 12.5%. (g) High-magnification SEM image of the hydrogel with 10% ionic liquid. (h) EDS elemental mapping images of C, N, O, and S for the hydrogel with 10% ionic liquid.

FTIR and XPS analyses further confirm the successful incorporation of various components into the PPEPSI hydrogel network. As shown in Figure S4a, increasing ionic liquid content leads to a decrease in the intensity and a slight shift of the broad -OH stretching band, indicating that $[\text{BMIM}][\text{DCA}]$ regulates the hydrogen-bonding interactions within the hydrogel network. The emergence of the $\text{C}\equiv\text{N}$ characteristic peak at approximately 2200 cm^{-1} confirms the successful introduction of the $[\text{DCA}]^-$ anion into the hydrogel network. In the full XPS spectra (Figure S4b), S 2p signals are observed in both PPEPS and PPEPSI hydrogels, indicating the successful incorporation of PEDOT:SSA into the hydrogel network. Furthermore, the high-resolution N 1s spectrum (Figure S4c) of PPEPSI exhibits the characteristic C-N^+ peak associated with the imidazolium cation, further confirming the successful incorporation of the ionic liquid.

Figure 4 illustrates the mechanical properties, electrical conductivity, thermal stability, and freeze resistance of the PPEPSI hydrogel. As shown in Figure 4a, all samples exhibit typical elastic mechanical behavior, with the stress increasing approximately linearly with strain before fracture. The

pristine PPEPS hydrogel without ionic liquid addition (0% group) exhibits a maximum tensile strength of 555 kPa, while its elongation at break is only 160%, indicating limited ductility. With increasing [BMIM][DCA] content, the tensile strength of the hydrogels gradually decreases, whereas the elongation at break increases significantly. The sample containing 10 wt% [BMIM][DCA] maintains a tensile strength of 436 kPa while achieving an elongation at break of 218%, demonstrating an optimal balance between strength and ductility. Although the 12.5 wt% group exhibits a further increase in elongation at break to 225%, its tensile strength decreases substantially to 257 kPa, which is unfavorable for maintaining structural stability. These changes in mechanical properties are mainly attributed to the plasticizing effect of [BMIM][DCA]. As small molecular additives, ionic liquids can penetrate between PVA/PAM polymer chains, weakening intermolecular interactions and increasing chain mobility, thereby improving the flexibility and extensibility of the hydrogel. However, the reduced intermolecular interactions simultaneously decrease the resistance against tensile deformation, resulting in a gradual reduction in tensile strength with increasing ionic liquid content. Considering the overall mechanical performance, 10 wt% [BMIM][DCA] is considered the optimal concentration, providing sufficient mechanical stability for flexible sensors under large deformation conditions. Furthermore, the PPEPSI hydrogel maintains structural integrity under various deformation modes, including stretching, bending, twisting, folding, cross-stretching, and knotting (Figure S5), demonstrating excellent flexibility and deformation adaptability.

To systematically investigate the influence of ionic liquid content on the electrical conductivity of PPEPSI hydrogels, EIS measurements were performed at room temperature using hydrogels containing different [BMIM][DCA] concentrations (0%, 5%, 7.5%, 10%, and 12.5%). The corresponding EIS spectra are shown in Figure 4b, and the conductivity values calculated according to Eq 1 are presented in Figure 4c. The PPEPS hydrogel without ionic liquid exhibits a relatively low conductivity of 10.01 mS/cm. With increasing [BMIM][DCA] content, the conductivity gradually increases and reaches a maximum value of 21.65 mS/cm at 10 wt% ionic liquid addition. However, further increasing the ionic liquid content to 12.5 wt% results in a decrease in conductivity, indicating that the influence of ionic liquid concentration on electrical conductivity does not follow a simple linear relationship but exhibits an optimal concentration range. The four-point probe measurements (Figure S6) show that the electronic conductivity of the hydrogel increases with IL content, rising from 0.086 mS/cm (0% IL) to a maximum of 0.146 mS/cm at 10 wt% IL, before slightly decreasing to 0.128 mS/cm at 12.5%. This trend indicates that [BMIM][DCA] may promote secondary doping of the PEDOT:SSA network up to an optimal loading, beyond which network disruption occurs. The TG/DTG curves (Figure 4d,e) demonstrate that the incorporation of PEDOT:SSA increases the residual carbon content of the hydrogel, suggesting enhanced thermal stability of the composite system. Meanwhile, the PPEPSI hydrogel maintains stable structural characteristics during thermal decomposition.

The DSC results in Figure 4f provide direct calorimetric evidence for the antifreezing mechanism. The PPE hydrogel exhibits a sharp crystallization exotherm at $-23.41\text{ }^{\circ}\text{C}$, indicating unambiguous bulk water freezing. Upon incorporation of PEDOT:SSA, the PPEPS hydrogel shows a significantly weakened and shifted exotherm at $-43.17\text{ }^{\circ}\text{C}$, demonstrating that a considerable fraction of water is prevented from crystallizing, although some bulk-like water remains capable of phase transition. Most importantly, the PPEPSI hydrogel shows no detectable crystallization exotherm across the entire tested temperature range (25 to $-80\text{ }^{\circ}\text{C}$). This complete absence of a phase-transition signal provides direct thermodynamic proof that ice formation is entirely suppressed in the PPEPSI system. The

disappearance of the crystallization peak may indicate that water molecules are strongly confined within the hydrogel network. Specifically, ethylene glycol forms extensive hydrogen bonds with water, and [BMIM][DCA] may disrupt the tetrahedral hydrogen-bond ordering required for ice nucleation through ion–dipole interactions. These synergistic interactions severely limit the translational mobility and orientational ordering of water molecules necessary for ice nucleation and growth, thereby kinetically and thermodynamically suppressing the liquid-to-solid transition. Figure 4g presents the changes in residual mass of the different hydrogels during storage. With increasing storage time, the residual mass of all samples gradually decreases; however, PPEPSI consistently exhibits the highest residual mass, whereas PVA/PAM/EG (PPE) shows the lowest value, with PPEPS located between them. These results indicate that PPEPSI possesses superior water retention capability. Improved water retention helps maintain a stable internal environment within the hydrogel and reduces structural changes caused by water migration under low-temperature conditions, which agrees well with the DSC results. Furthermore, as shown in Figure 4h, the PPEPSI hydrogel maintains excellent flexibility and stretchability at -20°C . It can withstand deformation processes such as knotting and stretching without obvious brittle fracture, further confirming its excellent freeze resistance.

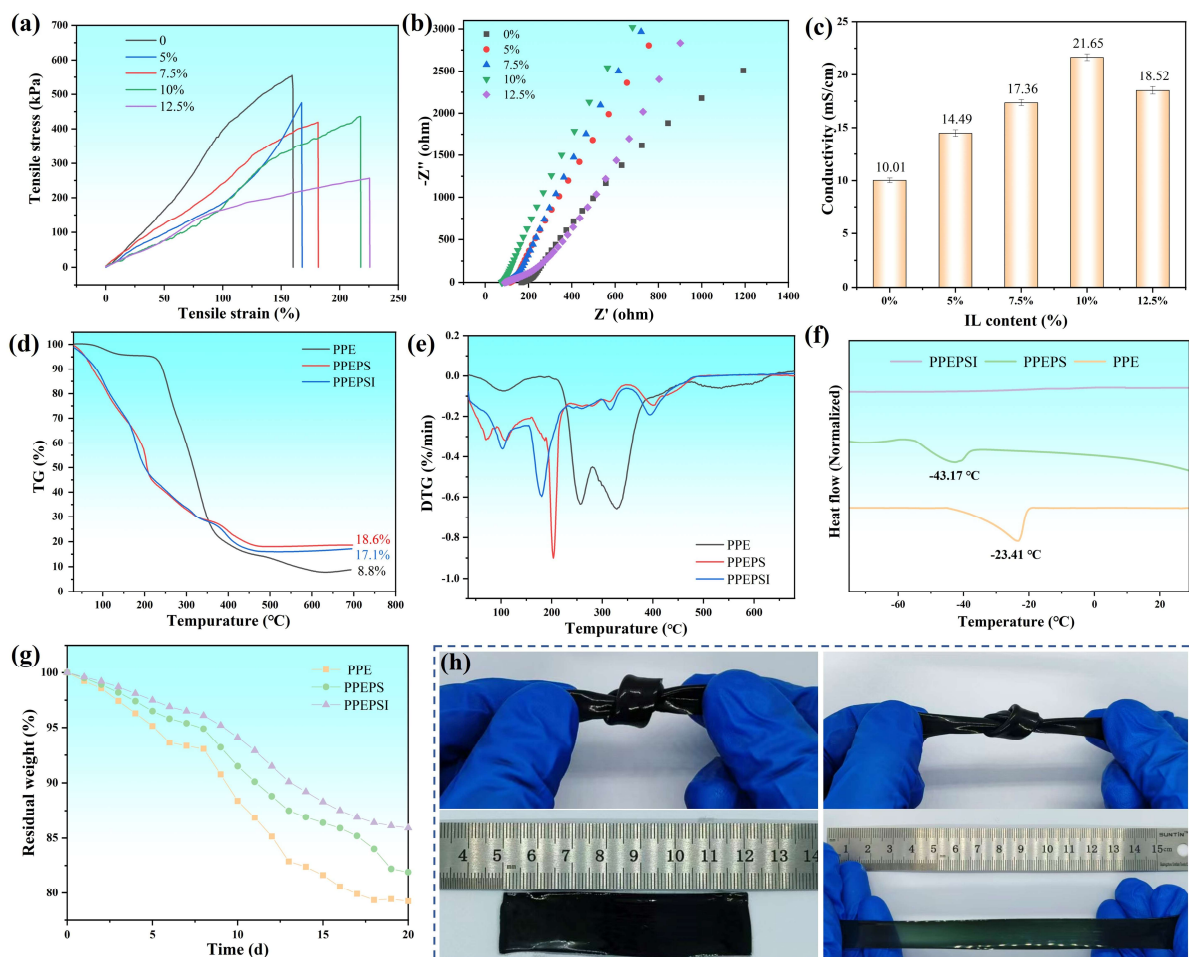


Figure 4. Comprehensive characterization of PPEPSI hydrogels: (a) tensile stress–strain curves of hydrogels with different ionic liquid contents; (b) Nyquist plots; (c) electrical conductivity; (d) TG curves; (e) DTG curves; (f) DSC curves; (g) water-retention behavior; and (h) demonstration of the antifreezing performance of PPEPSI hydrogel at -20°C .

In this work, PEDOT:SSA provides electronic conduction, with SSA serving as a dopant/dispersant to suppress PEDOT aggregation; EG depresses the freezing point via hydrogen bonding with water and acts as a plasticizer; the ionic liquid contributes ionic conduction and binds water molecules through ion–dipole interactions to prevent freezing, while also plasticizing the network; and the PVA/PAM double network provides the structural scaffold, where PVA crystalline domains ensure tensile strength, and the chemically cross-linked PAM network imparts elasticity and recovery. These distinct roles are supported by the control samples: PPE lacks conductivity and freeze resistance; PPEPS shows improved electronic conduction but still exhibits a DSC crystallization peak; and PPEPSI achieves the optimal synergy of high conductivity (21.65 mS/cm), no crystallization, and balanced mechanical properties.

3.3. Sensing performance of PPEPSI hydrogels

PPEPSI hydrogels exhibit stable resistive responses when used as flexible strain sensors. As shown in Figure 5a, the sensor exhibits a piecewise linear response over a wide strain range of 0–240%, with GF values of 1.82 (0–200%) and 3.06 (200–250%) in the corresponding strain regions, respectively, indicating its broad detection range and high sensitivity. Figure 5b shows that the sensor possesses rapid response and recovery times of 216 and 243 ms, respectively, enabling fast signal acquisition and feedback. Cyclic tensile tests demonstrate that the sensor generates stable and distinguishable resistance responses within the strain range of 100%–200% (Figure 5c). Furthermore, within the low strain range of 10%–80%, the sensor also exhibits stable and reproducible response behavior (Figure S7), demonstrating excellent strain resolution and response stability over a broad strain range.

Figure 5d presents the cyclic response of the sample subjected to 60% tensile strain at 25 °C (room temperature). No significant signal decay is observed, and stable periodic signals are well-maintained after 5000 s of continuous measurement, which verifies its outstanding cycling durability. The sensing mechanism is schematically illustrated in Figure 5e. During stretching, the polymer network undergoes deformation and orientation, increasing the distance between PEDOT:SSA conductive domains and disrupting partial conductive pathways, thereby resulting in increased resistance. Upon releasing the applied strain, the conductive network gradually recovers, and the resistance returns approximately to its initial value. PEDOT:SSA provides stable electronic conduction pathways, while [BMIM][DCA] improves ionic transport and enhances the flexibility of the hydrogel network. The synergistic effects of these components contribute to the excellent sensitivity and stability of the PPEPSI sensor.

Table 2 shows that most reported hydrogels excel in conductivity, strain range, or antifreezing, but rarely combine either with processability. For instance, PVA/PEDOT:PSS systems are printable yet non-antifreeze, whereas PVA/cellulose nanofibrils (CNF)/dimethyl sulfoxide (DMSO)/H₂O and polypyrrole (PPy)@CNF-PVA resist freezing but lack comparable conductivity or 3D printability. The PPEPSI hydrogel uniquely integrates high conductivity (2.165 S/m via EIS), freeze resistance, and 3D-printing capability. As shown in Table S2, its sensor achieves a gauge factor of 3.06 and a response time of 216 ms, outperforming most PEDOT-based counterparts in both sensitivity and speed, while maintaining a practical working range of 0–240%. Therefore, the as-fabricated PPEPSI hydrogel in this work exhibits comprehensive properties including favorable conductivity, antifreezing performance, sensing capability, and reprocessing feasibility.

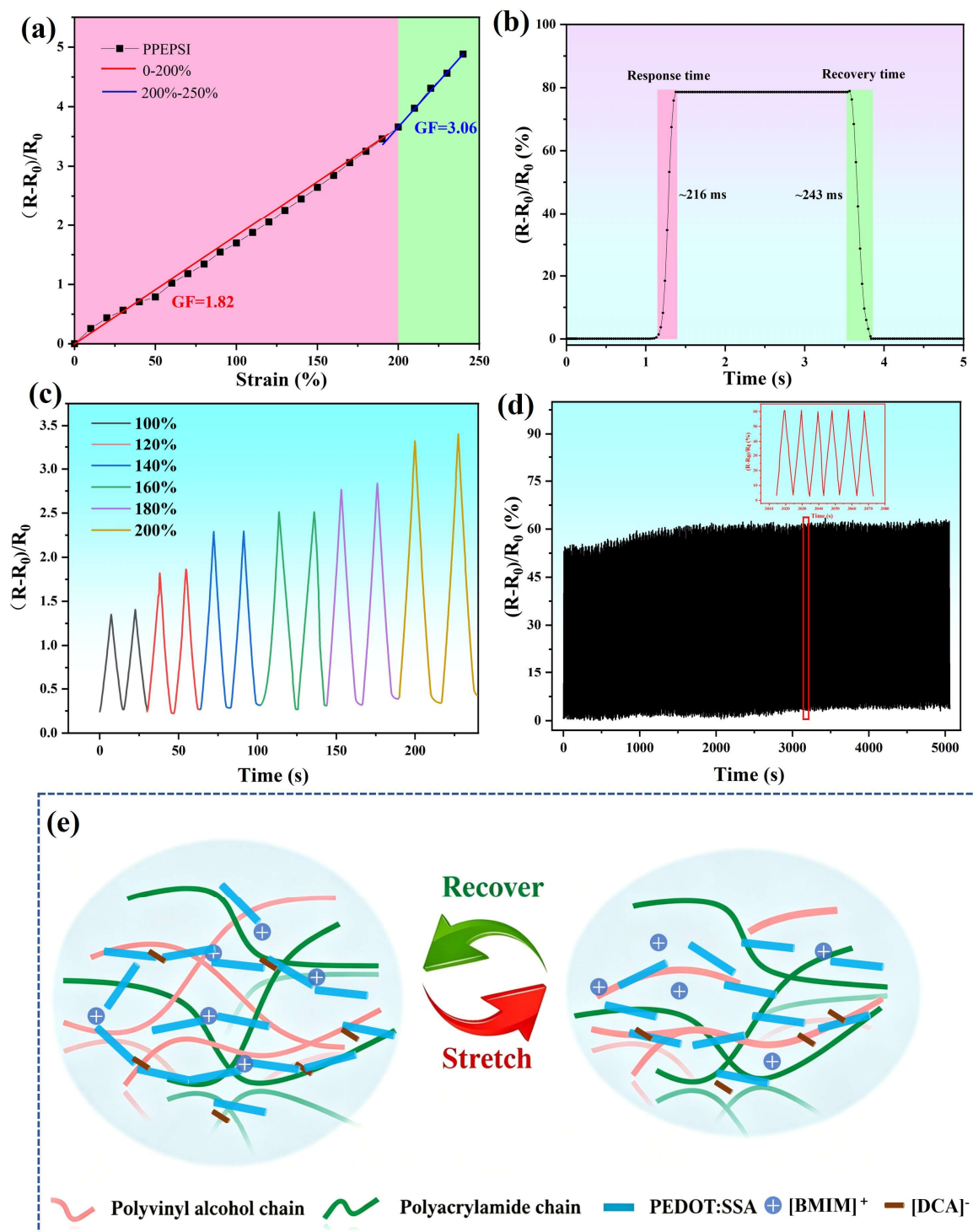


Figure 5. Strain-sensing performance and sensing mechanism of the PPEPSI hydrogel sensor: (a) relative resistance change as a function of strain and corresponding gauge factor (GF); (b) response and recovery time; (c) cyclic sensing responses under different strains; (d) long-term cyclic stability at 60% strain; and (e) schematic illustration of the strain-sensing mechanism of the PPEPSI hydrogel sensor.

Table 2. Comparison of basic physical properties of different hydrogels.

Materials	Conductivity	Strain range	Antifreezing behavior	Processability	Reference
PVA/PEDOT:PSS	0.4 S/m (four-point probe)	0–300%	/	All-3D printing	[68]
PVA/PEDOT:PSS/poly sulfobetaine methacrylate (PSBMA)	1.2 S/m (four-point probe)	0–349%	/	3D printing	[69]
CNF@PDA/PANI _(x) /PAM	0.257 S/m (inductance-capacitance-resistance (LCR) digital bridge instrument)	0–859.3%	/	---	[70]
PVA/CNF/DMSO/H ₂ O	3.2 S/m (EIS)	0–660%	Yes	---	[71]
PPy@CNF-PVA	0.031 S/m (EIS)	0–389%	Yes	---	[72]
PVA/DES	2.1 S/m (EIS)	0–390%	/	---	[73]
PVA/PAM/EG/PEDOT:SSA/IL	2.165 S/m (EIS), 0.0146 S/m (four-point probe)	0–240%	Yes	Can be 3D printed	Our work

To evaluate the practical application potential of PPEPSI hydrogels for wearable sensing, flexible sensors were attached to different human body locations, and a digital multimeter was used to record real-time resistance variations induced by mechanical deformation. As shown in Figure 6, the sensing performance of the PPEPSI hydrogel was systematically evaluated by monitoring human motions with different deformation amplitudes, including facial expressions, finger movements, palm bending, wrist motion, and elbow bending. Figure 6a,b demonstrates the capability of the sensor for human motion monitoring deformations. When volunteers puffed their cheeks or frowned, slight deformation of the skin surface induced structural changes in the internal conductive network of the hydrogel, resulting in regular resistance variations. These results indicate that the sensor possesses high sensitivity and reliable signal recovery capability for detecting weak physiological signals. The monitoring results of large-scale joint motions are presented in Figure 6c–f. When the fingers, palm, wrist, and elbow underwent bending or extension, the sensor generated clear and highly reproducible resistance responses. In particular, as shown in Figure 6c,f, when the bending angles of the fingers and elbows increased from 30° to 90°, the relative resistance change ($\Delta R/R_0$) exhibited a gradual increase. This angle-dependent response enables the sensor to distinguish different degrees of human motion. Furthermore, comparison of Figure 6d,e reveals that the sensor exhibits distinguishable resistance responses to different motion intensities at various body locations. All response curves maintain stable baselines and well-preserved signal waveforms during repeated deformation cycles, demonstrating the excellent stability and reliability of the PPEPSI hydrogel sensor for wearable motion monitoring.

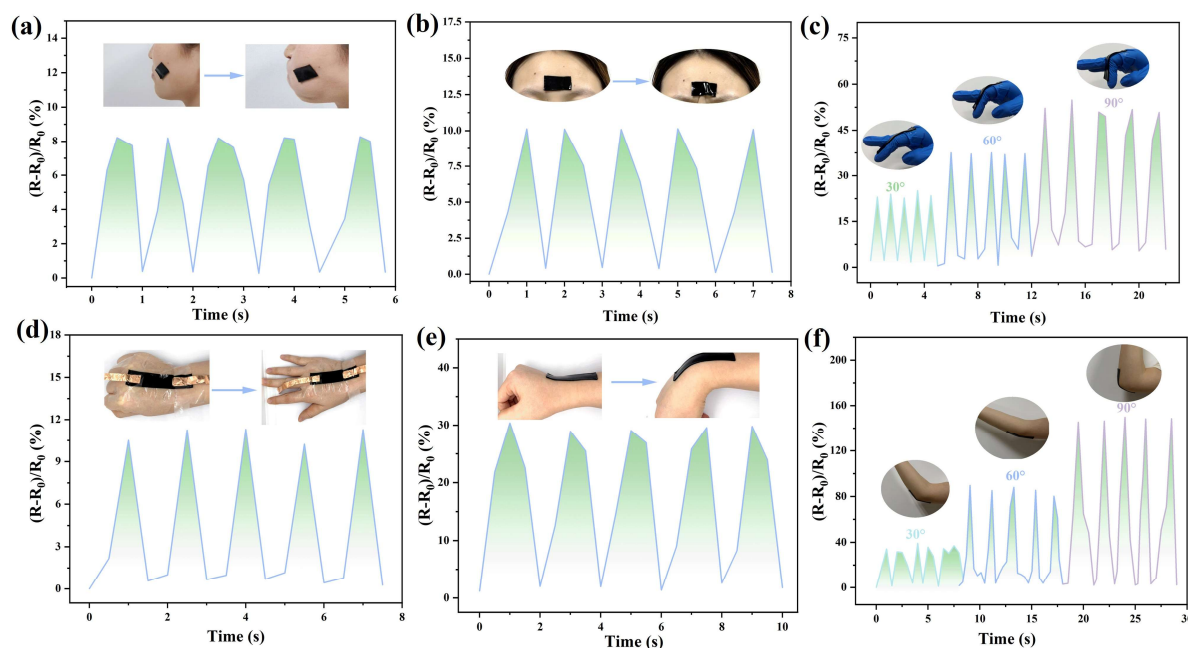


Figure 6. Real-time resistive response signals of PPEPSI hydrogel-based sensor for human motions at various body sites: (a) cheek puffing; (b) eyebrow frowning; (c) finger bending; (d) palm opening/closing; (e) wrist flexion; (f) elbow flexion.

In addition, the PPEPSI hydrogel demonstrates excellent handwriting signal differentiation. As shown in Figure 7a–d, the sensor successfully captures and distinguishes the characteristic resistance signals generated during the writing of the letters “b” and “d” as well as the words “Yes” and “No”. Figure 7a,b shows that writing different letters produces distinct resistance response patterns. The contact pressure applied during each stroke induces sharp negative resistance peaks, where the peak intensity reflects the applied pressure, while the peak width and waveform shape correspond to the duration and dynamic characteristics of the writing process. Due to differences in stroke sequences and deformation histories, the signal corresponding to “b” exhibits a more complex waveform, with an initial sharp decrease followed by signal fluctuations, whereas the signal for “d” presents a relatively symmetrical single-peak profile. These differences originate from the distinct mechanical deformation processes experienced by the hydrogel network during different writing motions, resulting in distinguishable resistance response patterns. Furthermore, Figure 7c,d demonstrates that the sensor can distinguish continuously written words. The signals generated by “Yes” and “No” consist of multiple characteristic peaks arranged in different temporal sequences. The differences in peak intervals and amplitude variations provide unique signal features for word identification. For example, “No” produces two characteristic peaks with specific temporal spacing, whereas “Yes” generates three peaks with different temporal relationships. These results indicate that the sensor is capable of distinguishing complex dynamic mechanical inputs with high temporal resolution. To evaluate the processability of the PPEPSI precursor for fabricating customized sensors, its rheological behavior was characterized in this study, and the results are presented in Figure 7e. As the shear rate increases, the viscosity of the precursor decreases continuously, exhibiting typical shear-thinning behavior. Such property enables the precursor to be extruded smoothly through the nozzle during direct-ink-writing (DIW) 3D printing, reduces processing resistance, and allows the fabrication of customized geometries without complicated

molds (Figure 7f). The shear-thinning behavior mainly originates from the reversible disentanglement of the PVA/PAM matrix under shear force. Overall, the excellent rheological properties of this precursor provide extra processing flexibility for the customized manufacturing of sensors with complex shapes, demonstrating its practical application potential in the field of flexible electronics. To investigate the remoldability of the PPEPSI hydrogel, a heating-dissolution and low-temperature reconstruction strategy was adopted, and the results are shown in Figure S8. The pristine PPEPSI hydrogel exhibited a stable bulk state. After being heated in a water bath at 90 °C for 5 min, the sample transformed into a homogeneous flowing state, indicating that its internal network structure dissociated under thermal treatment. Subsequently, the fluid-state sample was kept static at −20 °C for 8 h. The system regenerated into a hydrogel and could be reshaped into the characters “XUT”, demonstrating the excellent reprocessing feasibility of this hydrogel. The amplitude-sweep results (Figure S9) show that the storage modulus (G') of the PPEPSI precursor is consistently higher than its loss modulus (G'') at low shear stresses, indicating favorable self-supporting property and shape-retaining capability of the material under static conditions. When the shear stress rises to approximately 100 Pa, (G') intersects with (G'') at the flow point. This suggests that the material can be extruded smoothly under high shear. After extrusion, the material rapidly regains a high modulus upon stress release.

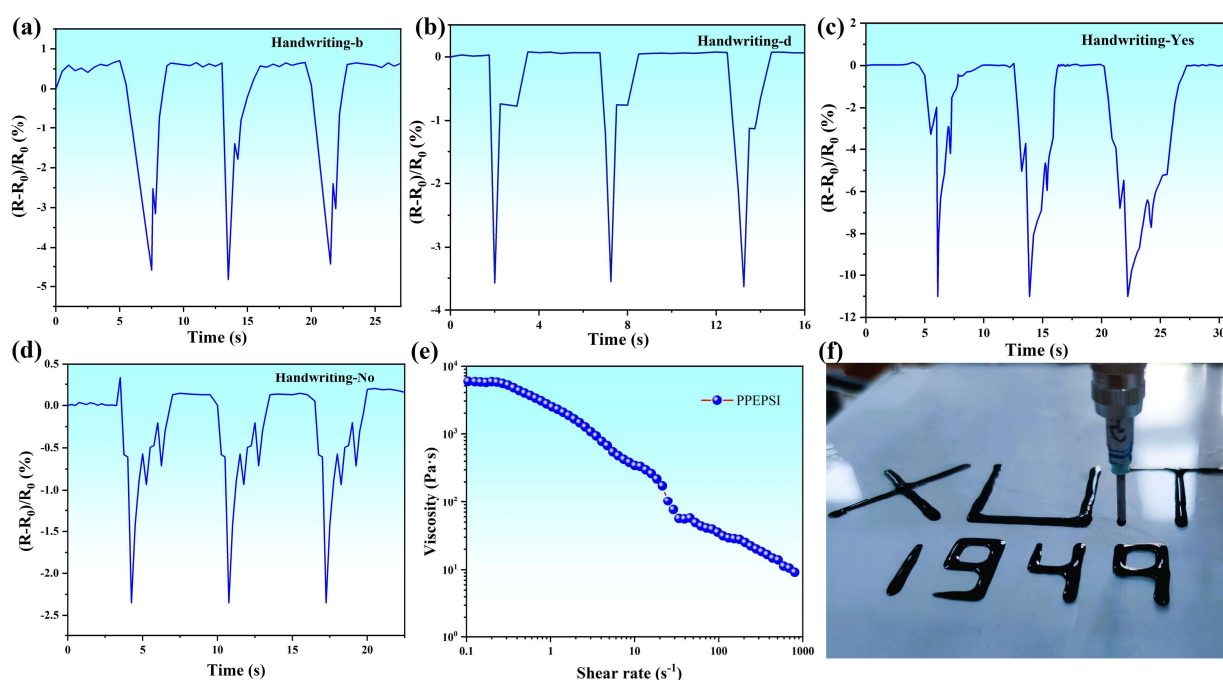


Figure 7. Human–machine interaction and 3D-printing performance of PPEPSI hydrogels: (a) resistance response signal for handwriting letter “b”; (b) resistance response signal for handwriting letter “d”; (c) resistance response signal for handwriting word “Yes”; (d) resistance response signal for handwriting word “No”; (e) viscosity–shear rate curve of the PPEPSI precursor; and (f) representative 3D-printed patterns fabricated from the PPEPSI precursor.

4. Conclusions

In this study, SSA was employed as both a dopant and dispersing agent to successfully prepare a PEDOT:SSA conductive composite. Furthermore, a PPEPSI double-network conductive hydrogel was constructed, integrating electrical conductivity, freeze resistance, and mechanical flexibility. The incorporation of SSA effectively improved the dispersion stability of PEDOT and enhanced its electrical conductivity, while the ionic liquid further regulated the conductive network structure and improved the low-temperature tolerance of the hydrogel. The resulting PPEPSI hydrogel exhibited excellent mechanical properties, electrical conductivity, and water retention capability, maintaining good flexibility and stable electrical performance even at $-20\text{ }^{\circ}\text{C}$. Moreover, the PPEPSI hydrogel-based sensor exhibited a wide strain sensing range (0–240%), high sensitivity ($GF = 1.82\text{--}3.06$), rapid response and recovery times (216/243 ms), and excellent cyclic durability, enabling applications in human motion monitoring and handwriting trajectory recognition. In addition, the hydrogel precursor solution showed pronounced shear-thinning behavior and good 3D printing capability, allowing the fabrication of complex structures. These findings demonstrate that PPEPSI conductive hydrogels have promising potential for applications in flexible electronics, wearable sensors, intelligent interactive wearable electronics, and 3D-printed functional devices. The superior dispersion stability and biocompatibility of SSA, combined with the intrinsic antifreeze properties of the PPEPSI network, suggest potential for long-term wearable implants and extreme-environment sensors. Future work will focus on the upper-limit performance of PEDOT:SSA and its application potential in extreme environments.

Use of AI tools declaration

The authors declare they have not used Artificial Intelligence (AI) tools in the creation of this article.

Acknowledgements

This work was supported by the Shaanxi Provincial Department of Education (Grant No. 22JY046), the Natural Science Basic Research Program of Shaanxi Province (Grant No. 2025JC-YBQN-656), the Scientific Research Program Funded by Shaanxi Provincial Education Department (Grant No. 24JK0563), the Doctoral Starting Foundation of Xi'an University of Technology (Grant No. 101-451124001), the International Cooperation Project of Xi'an University of Technology (Grant No. 101-256172504), and the Key R&D Program (General Project) of Shaanxi Provincial Department of Science and Technology (Grant No. 2026NC-YBXM-440).

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Conflict of interest

The authors declare no conflict of interest.

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