

Conductive Polyaniline Hydrogel Featuring High Toughness and Low Hysteresis

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Electronic Supplementary Information

Abstract Polyaniline (PANI) hydrogels have a wide range of applications in artificial skin, flexible robotics, and movement monitoring. Nevertheless, limited by the modulus mismatch between rigid PANI and the soft hydrogel matrix, the high strength and toughness of the PANI hydrogel are mutually exclusive. Although the introduction of sacrificial bonds into the hydrogel network can alleviate this contradiction to a certain extent, it always causes pronounced energy hysteresis during hydrogel deformation. Inspired by the energy storage and release of macroscopic springs, in this work, we propose a molecular entanglement approach for the fabrication of PANI hydrogels featuring high toughness and low hysteresis, where flexible poly(ethylene glycol) (PEG) is entangled with chemically cross-linked poly(acrylic acid) (PAA) as a hydrogel matrix, and rigid PANI as a conductive filler. The resultant PAA/PEG/PANI hydrogel exhibited high mechanical properties (fracture strength of 0.75 MPa and toughness of 4.81 MJ·m⁻³) and a low energy dissipation ratio (28.2% when stretching to 300%). Moreover, the PAA/PEG/PANI hydrogel possesses a good electrical response to external forces and can be employed as a strain sensor to monitor human joint movements by producing specific electrical signals. This work provides a straightforward strategy for preparing tough conductive PANI hydrogels with low hysteresis, showing potential for the development of healthcare devices.

Keywords Polyaniline hydrogel; Low hysteresis; High toughness; Physical entanglement

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INTRODUCTION

Polyaniline (PANI) hydrogel, consisting of PANI fillers and a hydrogel matrix, is a soft material that has a range of applications in artificial skin, flexible robotics, and movement monitoring.^[1,2] Owing to the modulus mismatch between rigid PANI and the soft hydrogel matrix, the high strength and toughness of PANI hydrogels are mutually exclusive.^[3,4] One of the proven strategies to alleviate this contradiction is the introduction of sacrificial bonds, as they are able to optimize interfacial interactions between fillers and the matrix, such as the modification of dopant acids in the polymerization of PANI,^[5] and the regulation of network structures of the hydrogel.^[6] Nevertheless, these sacrificial bonds, in turn, cause pronounced energy hysteresis during hydrogel deformation, causing the toughness gained from hysteresis to be ineffective in cyclic loading. The fracture

toughness of PANI hydrogels is expressed as $\Gamma = \Gamma_0 + \Gamma_D$, where Γ_0 refers to the intrinsic fracture toughness (1–100 J·m⁻²), and Γ_D represents the fracture toughness contributed by sacrificial bonds, which can be up to 1000 J·m⁻².^[7–11] Therefore, it is of great significance to develop PANI hydrogels with low hysteresis.

Like macroscopic springs, flexible macromolecules are capable of switching their conformations in response to external forces by undergoing mechanical motion.^[12] Under an applied force, the bond length, bond angle, and/or dihedral angle of flexible macromolecules are perturbed from equilibrium with energy storage. Once the force is released, they would prefer to return to the equilibrium conformation if no transition state is reached.^[13] This delicate process has inspired researchers to fabricate tough hydrogels with low hysteresis by incorporating topologically adjustable networks, where mechanical energy can be absorbed, stored, and transmitted through the conversion of molecular conformations under external forces.^[14–16] For example, Zhang *et al.*^[17] utilized the molecular spring of helical peptide chains to fabricate tough hydrogels. During consecutive loading-unloading cycles, mechanical energy is absorbed and stored through the deformation of the helical peptide chains, leading to extra-small hysteresis loops. Suo *et al.*^[18] deployed chain entan-

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lements instead of chemical/physical crosslinking to construct tough hydrogels. Because these entanglements enabled the tension transmission of the polymer chains to other chains, the hydrogel featured the integration of high toughness and negligible hysteresis. By designing topological networks with adjustable molecular conformations, the simultaneous realization of the high toughness and low hysteresis of PANi hydrogels will be feasible.

In this study, we proposed a molecular entanglement strategy to construct a tough conductive PANi hydrogel with low hysteresis, as illustrated in Fig. 1. Owing to the good molecular compatibility between poly(ethylene glycol) (PEG) and poly(acrylic acid) (PAA), their topological entanglement is beneficial for the mechanical performance.^[19] Upon loading, the chemically cross-linked network of PAA serves as a framework to stabilize the hydrogel, whereas PEG entangles with PAA to dissipate energy through topological conformation change, offering surplus physical cross-linking points for strengthening the hydrogel while maintaining low hysteresis. Meanwhile, the presence of conductive PANi fillers further transferred and dispersed stress, enhancing the fracture strength and modulus of the hydrogel. The resulting PAA/PEG/PANi hydrogel showed a fracture strength of 0.75 MPa, elongation at break of 910%, and toughness of 4.81 MJ·m⁻³. More importantly, the energy dissipation ratio remained as low as 28.2% when stretched to 300%. This low hysteresis endows the PAA/PEG/PANi hydrogel with a good electrical response to external forces, which can be used as a strain sensor to monitor the movement of human joints.

EXPERIMENTAL

Synthesis of PAA/PEG Hydrogel

Initially, acrylic acid (AA, 1.5 g) and poly(ethylene glycol) (PEG, $M_n=4000$ Da, 1 g) were dissolved in deionized water (8.5 mL), and then was heated at 60 °C to form a uniform and transpar-

ent aqueous solution. Subsequently, ammonium persulfate (APS, 95 μL , 2.19 $\text{mmol}\cdot\text{mL}^{-1}$) and *N,N*-methylene diacrylamide (MBA, 33 μL , 0.32 $\text{mmol}\cdot\text{mL}^{-1}$) were added at room temperature. After being placed in a mold, the aqueous solution was polymerization at 60 $^{\circ}\text{C}$, and the PAA/PEG hydrogel was obtained after 6 h. Notably, the acquired hydrogel was immersed in deionized water for 24 h to remove the initiator before testing for various purposes.

Synthesis of PAA/PEG/PANi Hydrogel

Initially, the PAA/PEG hydrogel was immersed in an aniline solution (0.1 mol/L) in the presence of acetic acid. Subsequently, APS with an equal molar content of aniline was added at room temperature and then transferred to an environment at 5 °C for *in situ* polymerization for 48 h. After soaking in deionized water for 24 h to remove unreacted monomer and initiator, the target PAA/PEG/PANi hydrogel was obtained.

The information of “Materials” and “Characterizations” are detailed in the electronic supplementary information (ESI).

RESULTS AND DISCUSSION

Fig. 1 shows the fabrication process of the conductive PAA/PEG/PANi hydrogel. Prior to the investigation of the PAA/PEG/PANi hydrogel, the effect of PEG on the mechanical properties of the PAA/PEG hydrogel was explored by changing the mass ratio of PEG to PAA from 0 to 1/3, 2/3, and 1, which were named PAA/PEG₀, PAA/PEG_{1/3}, PAA/PEG_{2/3}, and PAA/PEG_{3/3}, respectively. Tensile tests under a stretching speed of 20 mm·min⁻¹ showed that the fracture strengths were 27.71, 40.61, 89.41, and 47.11 kPa for PAA/PEG₀, PAA/PEG_{1/3}, PAA/PEG_{2/3}, and PAA/PEG_{3/3} hydrogels, respectively, accompanied by an elongation at break of 1124% to 773%, 731%, and 430%, respectively (Fig. 2a). By integrating the stress-strain curves, their toughness values were calculated and are summarized in Fig. S1 (in ESI), where the PAA/PEG_{2/3} hydrogel pos-

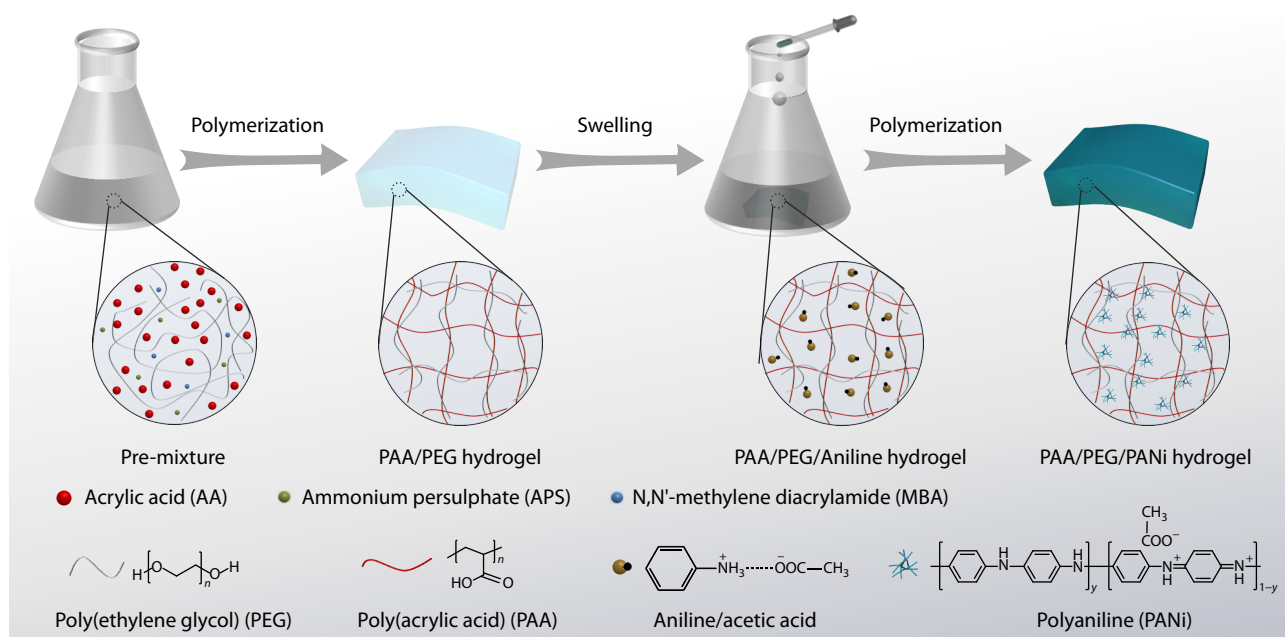


Fig. 1 Schematic illustration of the preparation of conductive PAA/PEG/PANi hydrogel.

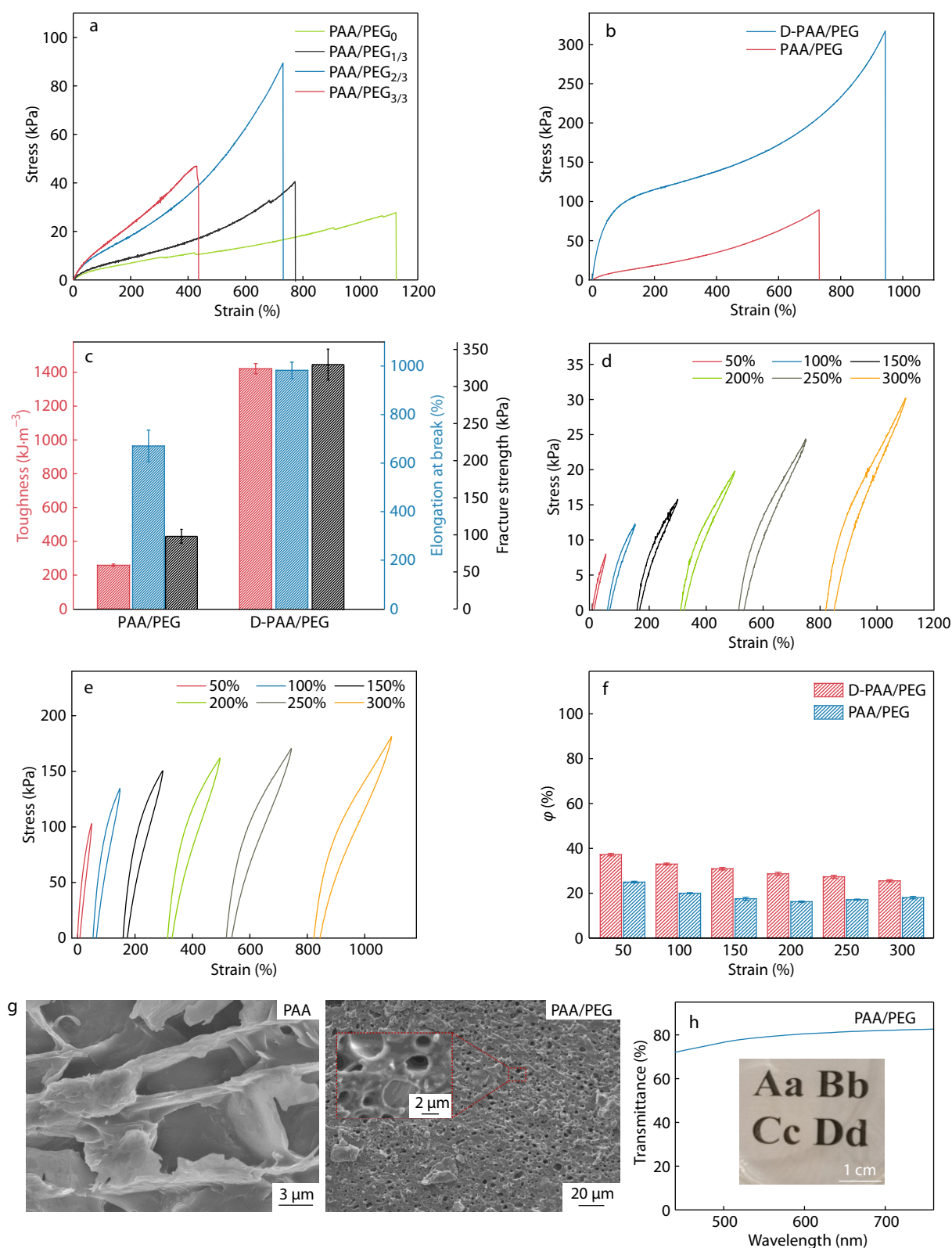


Fig. 2 (a) Tensile stress-strain curves of PAA/PEG hydrogels; (b) Tensile stress-strain curves of PAA/PEG and D-PAA/PEG, and (c) their corresponding values of toughness, elongation at break, and fracture strength. Continuous cyclic stretching-releasing tests of (d) PAA/PEG and (e) D-PAA/PEG with the strain from 50% to 300% in an increment of 50%; (f) Energy dissipation ratio (ϕ) of PAA/PEG and D-PAA/PEG calculated from their cyclic stretching-releasing tests; (g) SEM images of PAA and PAA/PEG hydrogels; (h) UV-Vis spectrum of PAA/PEG hydrogel. Inset demonstrates the transmittance of PAA/PEG hydrogel.

essed the highest toughness ($263.41 \text{ kJ}\cdot\text{m}^{-3}$). As a result, the PAA/PEG_{2/3} hydrogel was selected for the following investigation, and was further re-named as PAA/PEG for easy discrimination.

To better understand the role of PEG in the mechanical performance, the PAA/PEG hydrogel was dehydrated to form a D-PAA/PEG gel for tensile tests. Compared with the PAA/PEG hydrogel, D-PAA/PEG exhibited a higher fracture strength (331.31 kPa), elongation at break (982%), and toughness ($1.42 \text{ MJ}\cdot\text{m}^{-3}$) (Figs. 2b and 2c), indicating that PEG could entangle with PAA to strengthen the hydrogel. Cyclic stretching-releasing tests showed that the unloading curves of the PAA/PEG hydrogel were almost superimposable on the loading curves within a wide strain range (50% – 300%), displaying minor hysteresis loops (Fig. 2d). The energy dissipation ratios (ϕ) were calculated to be 24.9% , 20.0% , 17.5% , 16.2% , 17.1% , and 18.0% at strains of 50% , 100% , 150% , 200% , 250% , and 300% , respectively, which are comparable to other reported gels

with low hysteresis (Fig. 2f and Table S1 in ESI). Not surprisingly, the ϕ of D-PAA/PEG remained low, that is, 37.2% at 50% , 32.9% at 100% , 30.9% at 150% , 28.7% at 200% , 27.4% at 250% , and 25.5% at 300% (Figs. 2e and 2f), indicating that the physical entanglements of PEG with PAA effectively stored and released energy via the transformation of the molecular configuration, consequently relieving the hysteresis of the inner structures.^[20,21] Because of the topological entanglements for shape retention, the PAA/PEG hydrogel exhibited a good anti-swelling capacity (Fig. S2 in ESI). As displayed in the swelling curves, the swelling ratio of the PAA/PEG hydrogel was only 199% , which was much lower than that of the PAA hydrogel (Fig. S3 in ESI). Scanning electron microscope (SEM) images further proved the denser microstructures of PAA/PEG hydrogel (Fig. 2g), where the pore size was around $2.89 \mu\text{m}$, much smaller than that of PAA hydrogel (Fig. S4 in ESI), again revealing the presence of topological entanglements within the hydrogel network. Certainly, this entangle-

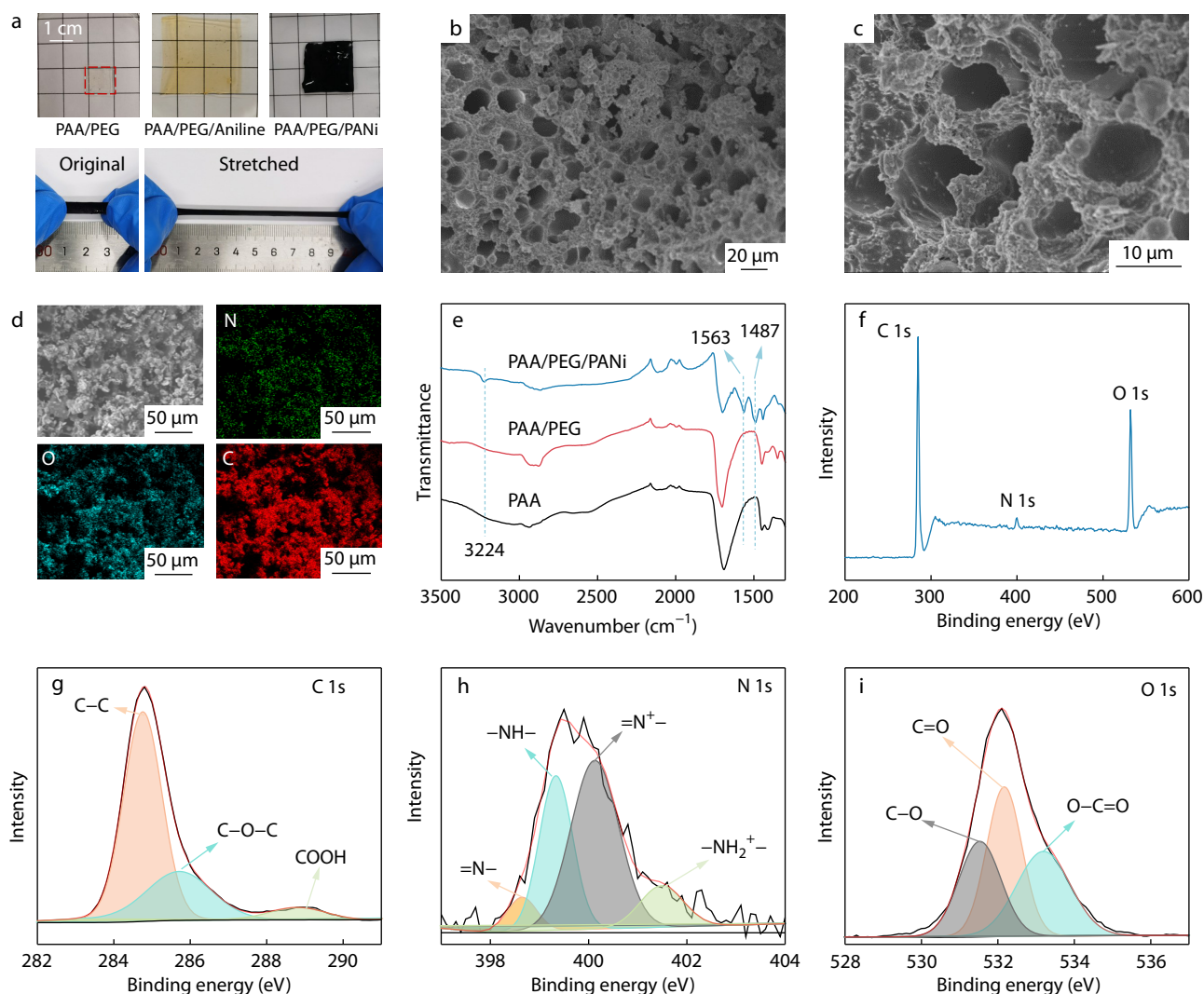


Fig. 3 (a) Digital photos of PAA/PEG, PAA/PEG/Aniline, and PAA/PEG/PANI hydrogel at original and stretchable state; (b) SEM image of PAA/PEG/PANI hydrogel and (c) its magnified vision; (d) SEM-mapping of PAA/PEG/PANI hydrogel with element distribution of O, C, and N; (e) FTIR spectra of PAA, PAA/PEG, PAA/PEG/PANI hydrogels in wavenumber range from 1300 cm^{-1} to 3500 cm^{-1} ; (f) XPS spectra of PAA/PEG/PANI hydrogel, and the corresponding high-resolution spectroscopy of (g) C 1s, (h) N 1s, and (i) O 1s.

ment did not influence the transparency of the PAA/PEG hydrogel, where the transmittance remained at approximately 80% in the wavelength range of 380–780 nm (Fig. 2h).

In situ polymerization of aniline was employed to introduce PANi into the PAA/PEG hydrogel. Initially, the PAA/PEG hydrogel was immersed in an aniline solution protonated by acetic acid to obtain a light-yellow PAA/PEG/Aniline hydrogel (Fig. 3a). After the addition of APS to initiate *in situ* polymerization of pre-introduced aniline, a dark green stretchable hydrogel was formed, demonstrating the successful loading of conductive PANi. SEM images showed that the pore surface of the hydrogel changed from smooth to puckered after introducing PANi (Figs. 3b and 3c), with the appropriate deposition of PANi (Fig. S5 in ESI). Moreover, the distribution of C, N, and O elements was clearly observed (Fig. 3d), and their relative contents were 69.7%, 7.4%, and 22.9%, respectively (Fig. S6 in ESI). In particular, the presence of N confirms the successful introduction of PANi.^[22,23] Fourier transform infrared

spectroscopy (FTIR) afforded similar results, in which the bands at 3224, 1563 and 1487 cm^{-1} corresponded to the stretching vibrations of the N—H bond, quinone, and benzene rings of PANi, respectively (Fig. 3e).^[24]

X-ray photoelectron spectroscopy (XPS) was utilized as a deeper characterization to verify the elemental composition and structure of PAA/PEG/PANi hydrogel. The results showed that C 1s, N 1s, and O 1s were located at 285.0, 400.0 and 532.0 eV, respectively (Fig. 3f). For C 1s region, it could be divided into three electronic states at 284.8 (C—C), 285.7 (C—O—C), and 288.8 eV (COOH) (Fig. 3g), which proved the simultaneous existence of PAA and PEG.^[25] The N 1s region was decomposed into four peaks at 398.7, (=N—), 399.3 (—NH—), 400.1 (=NH⁺—), and 401.5 eV (—NH₂⁺—), respectively, verifying the conductive emerald salt form of PANi (Fig. 3h).^[26] Moreover, the presence of C—O, C=O, and O—C=O at 532.2, 533.2, and 531.5 eV in high-resolution O 1s spectrum indicated that the presence of conductive PANi did not

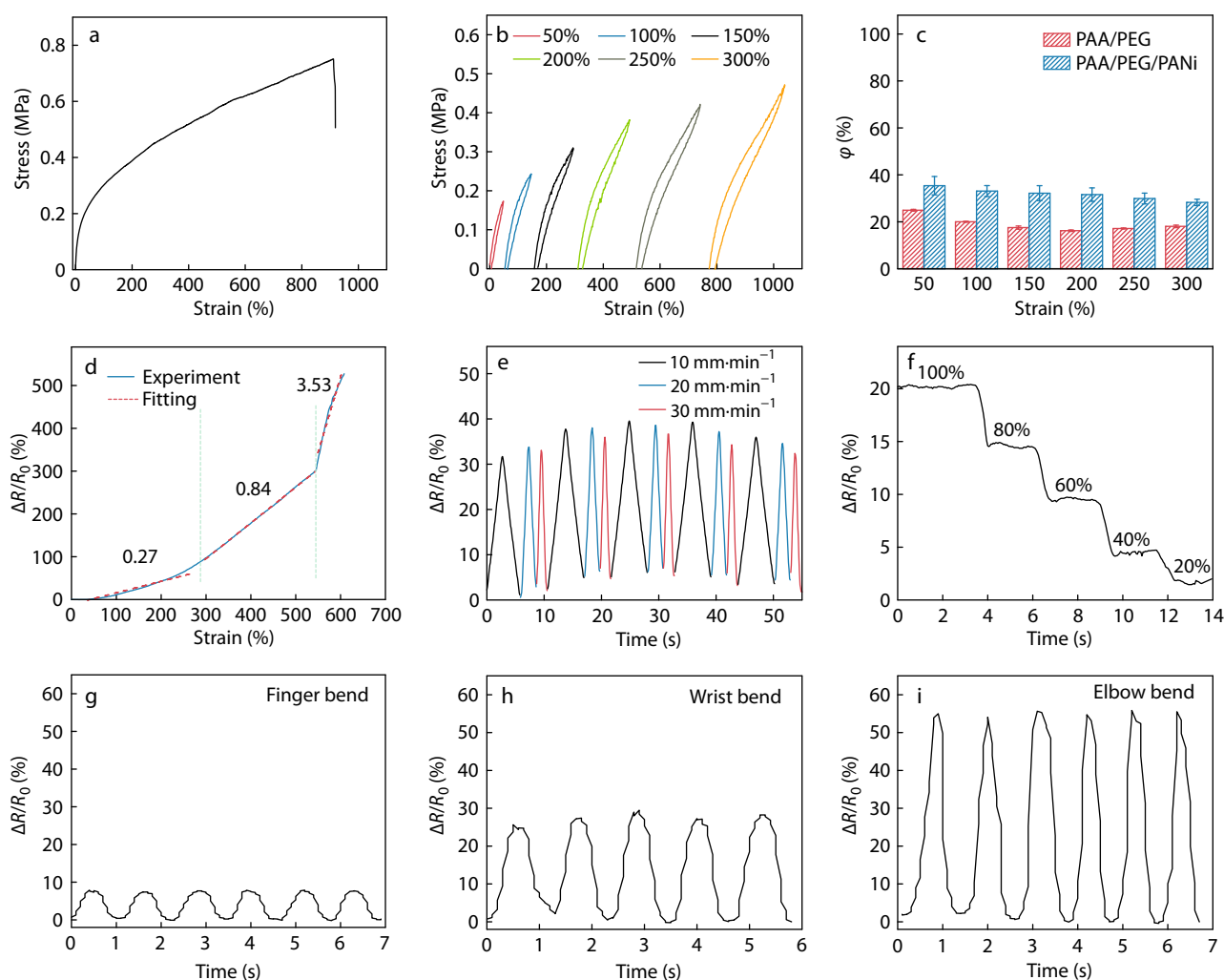


Fig. 4 (a) Tensile stress-strain curves of PAA/PEG/PANi hydrogel; (b) Continuous cyclic stretching-releasing tests of PAA/PEG/PANi hydrogel with the strain from 50% to 300% in an increment of 50%; (c) Energy dissipation ratio (ϕ) of PAA/PEG and PAA/PEG/PANi hydrogel calculated from their cyclic stretching-releasing tests; (d) Relative resistance variation under monodirectional strain (0%–630%); (e) Electrical response of PAA/PEG/PANi hydrogel under 200% strain at stretching rates of 10, 20, and 30 $\text{mm}\cdot\text{min}^{-1}$, respectively; (f) The variation of electrical signal of PAA/PEG/PANi hydrogel under gradient external deformation. Sensing performance in monitoring electrical signal variations to human joint movements like (g) finger bend, (h) wrist bend, and (i) elbow bend.

destroy the original structure of PAA/PEG network (Fig. 3i).^[27]

Tensile tests of the PAA/PEG/PANi hydrogel were performed using rectangular specimens with dimensions of 1 mm × 5 mm × 40 mm at a stretching speed of 20 mm·min⁻¹. As displayed in Fig. 4(a) and Fig. S7 (in ESI), the PAA/PEG/PANi hydrogel exhibited high mechanical performance, with a fracture strength of 0.75 MPa, elongation at break of 910%, and toughness of 4.81 MJ·m⁻³. When the loading amount of PANi decreased, its mechanical performance was significantly weakened (Fig. S8 in ESI). As expected, the PAA/PEG/PANi hydrogel possessed low hysteresis in continuous cyclic stretching-releasing tests (Fig. 4b) and ϕ was measured to be 35.4%, 33.1%, 32.2%, 31.6%, 29.9%, and 28.2% at strains of 50%, 100%, 150%, 200%, 250%, and 300%, respectively, which were slightly higher than those of the PAA/PEG hydrogel under the same strain (Fig. 4c). In addition, the PAA/PEG/PANi hydrogel exhibited appropriate stability in the time-sweep test (Fig. S9 in ESI), indicating its feasibility for sensing performance. At the same time, the PAA/PEG/PANi hydrogel exhibited a stepped response during stretching, where the gauge factor (GF), defined as the ratio of the relative resistance change rate to the applied strain, was calculated to be 0.27 within 0%–300% strain, 0.84 within 300%–550% strain, and 3.53 within 550%–620% strain (Fig. 4d). In contrast, the GF of the PAA/PEG hydrogel was much lower (0.08) owing to the lack of conductive PANi (Fig. S10 in ESI), which strongly indicated the specific role of PANi in enhancing the strain-sensing capability. When the PAA/PEG/PANi hydrogel was stretched to a fixed strain of 200% at tensile rates of 10, 20, and 30 mm·min⁻¹, the electrical resistance variation varied from 31.7% to 39.6% in small increments with different response times (Fig. 4e). Upon stretching to a fixed strain of 30% for 100 times, its electrical resistance variation remained at approximately 3% (Fig. S11 in ESI), demonstrating the good stability and durability of the PAA/PEG/PANi hydrogel as a strain sensor. Moreover, in the strain-electrical retention experiment, the electrical resistance variation decreased steadily from 20% at a strain of 100% to 2% at a strain of 20% (Fig. 4f), further proving the sensor stability. Benefiting from the above good comprehensive performance, the PAA/PEG/PANi hydrogel can be used to monitor the movements of human joints by producing specific electrical signals, such as bending of the finger, wrist, and elbow (Figs. 4g–4i), showing great potential for healthcare devices. Compared to other conductive hydrogels, such as poly(3,4-ethylenedioxythiophene) (PEDOT) and polypyrrole (PPy) hydrogels,^[28–30] the PAA/PEG/PANi hydrogel exhibited unique electrical performance and low mechanical hysteresis, which guaranteed their latent competitiveness.

CONCLUSIONS

In summary, using a molecular entanglement strategy, we successfully prepared a tough conductive PAA/PEG/PANi hydrogel featuring low hysteresis. Upon loading, the chemically cross-linked PAA served as a framework to stabilize the hydrogel network, the rigid PANi fillers transferred and dispersed stress, and the entanglements of PEG with PAA offered surplus physical

cross-linking points for strengthening the hydrogel by maintaining low hysteresis. As a result, PAA/PEG/PANi hydrogel exhibited good mechanical performance with a fracture strength of 0.75 MPa, elongation at break of 910%, toughness of 4.81 MJ·m⁻³, and energy dissipation ratio as low as 28.2% when stretching to 300%. Moreover, the negligible hysteresis effectively prevented the destruction of the electron transportation pathway in the hydrogel and consequently staved off response retardation, with GF of 0.27 (strain of 0%–300%), 0.84 (strain of 300%–550%), and 3.53 (strain of 550%–620%). As a strain sensor, the PAA/PEG/PANi hydrogel could monitor the movements of human joints by producing specific electrical signals. This study provides a simple yet effective approach to fabricate conductive hydrogels with high toughness and low hysteresis, showing potential for the development of healthcare devices.

Conflict of Interests

The authors declare no interest conflict.

Electronic Supplementary Information

Electronic supplementary information (ESI) is available free of charge in the online version of this article at <http://doi.org/10.1007/s10118-025-3283-0>.

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