Stretchable, sensitive, and environment-tolerant ionic conductive organohydrogel reinforced with cellulose nanofibers for human motion monitoring

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Abstract Conductive hydrogel (CH) strain sensors have made significant progress in wearable electronic products in recent years. However, the use of aqueous solvents as the dispersion medium in CHs largely limits the scope of applications of CHs and impedes the combination of the mechanical properties and ionic conductivity, which is urgently desired to be addressed. Herein, a simple one-pot preparation of anti-freezing, anti-drying ionic CHs with high stretchability (up to 869%), toughness (6.60 MJ/m³), and Young’s modulus (0.56 MPa) was proposed. These CHs consist of polyvinyl alcohol, tannic acid, and sodium chloride dispersed in a solvent consisting of glycerol and cellulose nanofiber suspension. The thus-synthesized CHs exhibit good ionic conductivity (~ 0.86 S/m) and strain sensitivity (gauge factor of 8.54). The organohydrogel possesses a sensitive strain sensing capability and a wide-working temperature range (−50 °C to 60 °C), and good stability (30 d in room-temperature) to detect human movement, such as large (joint movement) and subtle movements (voice in the throat). These advantages allow organohydrogel sensors to show great potential for electronic skin, personal healthcare, and flexible wearable devices.

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Introduction

Flexible wearable electronic devices based on conductive hydrogels (CHs) have been widely studied in recent years and used in the production of electronic skin, artificial intelligence, and human health monitoring (Lim et al. 2020; Zhang et al. 2020; Ye et al. 2020). Conventional CHs are mainly composed of water and susceptible to environmental factors. For instance, water molecules evaporate over time at room temperature or freeze to ice solids at temperatures below zero; alternatively, hydrogels tend to be dehydrated and become dry and hard at high temperatures (Ma et al. 2019). The hydrogel gradually loses its original mechanical properties, flexibility, and ionic conductivity during its service; its working temperature range can also be reduced (Yang et al. 2021b; Morelle et al. 2018).

Most conventional hydrogels are limited by the external use environments (He et al. 2020), low-volatility solvents ethylene glycol (Wei et al. 2020), glycerol (Gly) (Fang et al. 2020; Gu et al. 2020), and dimethyl sulfoxide (Ye et al. 2020), which are introduced into the gel to replace part of the water molecules. With this approach, the formation of low-temperature ice crystals and dehydration properties at high temperatures is effectively inhibited via strong hydrogen bonding interactions between organic solvents and water molecules (Liu et al. 2020; Hu et al. 2020; Lee et al. 2016). The strong hydrogen bonding improves the stability of the gel for its use in harsh environments and considerably slows down the drying rate. However, a reduction in the content of water molecules in the hydrogel system severely affects the electron conduction pathways and subsequently, the ionic conductivity of CHs. To enhance the conductivity of hydrogel sensors, conductive components such as polyaniline (Hu et al. 2018; Wang et al. 2018), polypyrrole (Chen et al. 2019b; Han et al. 2018b), carbon nanotubes (Han et al. 2018a; Cai et al. 2017; Jiao et al. 2021), graphene (Pan et al. 2020b; Alam et al. 2017), sodium chloride (Liu et al. 2020; Huang et al. 2020), and lithium chloride (Ge et al. 2021) are introduced into hydrogels. Owing to their simple, economical, and practical preparation and their inherent ductility and biocompatibility (Hu et al. 2020; Pan et al. 2020a; Shao et al. 2018), inorganic salt ion-based conductive hydrogels have shown great potential for applications in flexible wearable sensors. However, the wide application of ionic conductive hydrogels is hindered by the difficulty to strike a balance between the high conductivity and mechanical properties of ionic hydrogels, because the salting-out effect of electrolyte salts may disrupt the three-dimensional network of the hydrogel (Zhang et al. 2019; Yang and Yuan 2019). Numerous reports have shown that cellulose nanofibers (CNFs) enhance the mechanical properties of hydrogel systems (Jing et al. 2019; Zhu et al. 2021; Kong et al. 2018; Yang et al. 2021a). CNFs
consisting of $\beta$ (1→4) $\alpha$-glucose units exhibit a distinct chemical structure and the advantage of a large number of active hydroxyl sites, allowing more cross-linking sites and forming a more complex cross-linked network structure (Niu et al. 2021; Liu et al. 2021). Thus, CNFs are expected to further enhance the mechanical properties of ionic conductive hydrogels.

In this study, a simple one-pot process was used to prepare a highly stretchable (up to 869%), strong (up to 1.38 MPa), and tough (6.60 MJ/m$^3$) organohydrogels enhanced using cellulose nanofibrils. The organohydrogel was synthesized by mixing CNFs, tannic acid (TA), polyvinyl alcohol (PVA), and NaCl into a water-glycerol binary solvent, which also acted as the dispersion medium. Owing to the strong and abundant hydrogen bonds formed between water and glycerol, the environmental tolerance and long-term stability of the organohydrogel can accurately detect signals during human finger movement even after 30 d of storage and 12 h of storage in extremely harsh environments ($\sim$ 50 °C or 60 °C). The excellent mechanical properties of the organohydrogel are attributed not only to the complex cross-linked structure of the gel system but also to the CNF extracted from poplar wood powders, which acts as a reinforcing phase. The addition of NaCl to the hydrogel provides high ionic conductivity. The developed synthetic organohydrogel can address the concerns related to the mechanical strength, durability, ionic conductivity, and extreme environmental applicability of hydrogels, in addition to the high sensitivity and long-term stability of this hydrogel for detecting human motion. These advantages allow PVA/CNF/TA/Gly/NaCl organohydrogel sensors to show great potential for electronic skin, personal healthcare, and flexible wearable devices.

**Experimental section**

**Materials**

Polyvinyl alcohol-1799 (PVA), and tannic acid (TA) were purchased from Shanghai Aladdin Bio-Chem Technology Co. (Shanghai, China). NaCl and glycerin (Gly) were supplied by Tianjin Fuyu Fine Chemical Co. Ltd.

**Preparation of cellulose nanofibers (CNFs)**

CNFs were prepared by combining chemical pretreatment and strong ultrasonic treatment of poplar wood powder as previously described (Chen et al. 2011; Yue et al. 2021).

**Preparation of the PVA/CNF/TA/Gly/NaCl organohydrogel**

The hydrogel was prepared using the one-pot method. Firstly, 2.5 g PVA, 0.8 g TA, and NaCl of different qualities were added to a solution consisting of 10 g glycerol and 20 g CNF suspension (0.5 wt%, containing 0.1 g CNF) by magnetic stirring under 95 °C for 4 h. The hot solution underwent ultrasound treatment for 30 s to eliminate the gas in the solution before pouring it into the tailored mold at room temperature. Different PVA/CNF-x/TA/Gly/NaCl-y organohydrogels were obtained by varying the concentrations of the CNF suspension and the mass of NaCl (x and y represent the mass percentage ratios of CNF to PVA and NaCl to H$_2$O, respectively). The compositions of the hydrogels are listed in Table S1.

**Characterizations**

**Mechanical tests**

Tensile and compression tests of hydrogels were performed on a universal electronic machine tester (CMT5504, MTS system, China) equipped with a 2 kN loading cell. The hydrogel was cut into a dumbbell shape measuring 75 mm in length, 5 mm in width, and 1 mm in thickness. Tensile testing was conducted at 50 mm/min, and each sample was measured 7 times. The uniaxial compression testing was performed on cylindrical samples (20 mm in diameter and 15 mm in height) at a crosshead speed of 5 mm/min, and each sample was tested 5 times. Toughness was derived from the area under the stress–strain curve, and Young’s modulus was calculated from the initial slope of the stress–strain curve (between elongation $\sim$ 5% and $\sim$ 15%).
Fourier transform infrared spectroscopy

The chemical structure of the hydrogel samples was analyzed at room temperature in the 4000–600 cm\(^{-1}\) range by the Fourier transform infrared (FTIR, NICOLET 6700) spectroscopy.

Differential scanning calorimetry analysis

The antifreeze resistance of the PVA/CNF/TA/Gly/NaCl organohydrogel was analyzed by the differential scanning calorimetry (DSC TA Q20). The organohydrogel samples were cooled from room temperature to -70 °C at 5 °C/min under a nitrogen flow rate of 50 mL/min, kept for 5 min, and warmed to room temperature at the same rate.

Electrical measurements and organohydrogel sensor testing

The ionic conductivity of the organohydrogel was measured using the LCR digital bridge (TH2830, Changzhou, China). Conductivity (\(\sigma, \text{S/m}\)) was calculated using the following equation:

\[
\sigma = \frac{L}{R \times S}
\]

where \(L\) is the distance between the electrodes, \(R\) is the resistance of the sample, and \(S\) is the contact cross-sectional area of the sample.

The LCR digital bridge was used to record the real-time resistance changes under different strains at a voltage of 1 V and a scanning frequency of 1 kHz. The strain sensitivity of the organohydrogel (30 mm \(\times\) 5 mm \(\times\) 1 mm) was evaluated based on the gauge factor (GF), which is the ratio of the relative resistance change to the applied strain (\(\varepsilon\)).

\[
GF = \frac{(R - R_0)/R_0}{\varepsilon}
\]

where \(R\) is the resistance under the corresponding tensile strain, and \(R_0\) is the resistance in the original state.

Results and discussion

Synthesis of the organohydrogel

In this study, ionic conductive organohydrogels with desirable mechanical properties, excellent ionic conductivity, and superior water retention were prepared using a simple one-pot method. The schematic of the hydrogel preparation is presented in Fig. 1. The organohydrogel was prepared by mixing PVA, CNFs, TA, and NaCl homogeneously in the glycerol–water (mass ratio of 1:2) binary solvent under constant stirring. PVA was chosen as the main polymer network structure because of its excellent mechanical properties and elasticity. The rigid and negatively charged nanomaterial CNF was used as a reinforcement to further enhance the mechanical properties of the hydrogel. The introduction of TA containing high content of phenolic hydroxyl groups into hydrogels can effectively improve their tensile properties (Lin et al. 2020; Rahmani and Shojaei 2021). A strong and complex hydrogen bonding cross-linked network among PVA, TA, and CNF formed a good three-dimensional network framework for the hydrogel. NaCl was added as an electrolyte, exerting a salting-out effect to enhance the electrostatic interaction of the hydrogel (Ge et al. 2020). Moreover, ionic hydrogels composed of sodium salts were typically solvated viscoelastic flexible polymer network structures and can absorb large amounts of water molecules, which effectively provided channels for ion transport (Tao et al. 2017). To maintain the flexibility, ductility, and ionic conductivity of the hydrogel under extreme conditions, part of the water was replaced with glycerol to reduce the vapor pressure of water. Glycerin and water molecules consequently formed a strong hydrogen bond that effectively inhibited the formation of ice crystals and prevented the evaporation of water molecules (Liu et al. 2020). The binary solvent system composed of glycerin and water also introduced non-covalent crosslinks into the polymer system, enhancing the mechanical properties of the organohydrogel.

Mechanical properties of the organohydrogel

The mechanical properties of the organohydrogel were evaluated by typical tensile and compression tests. Both PVA/CNF/TA/Gly and PVA/CNF/TA/
Gly/NaCl hydrogels showed elastic behavior with high stress and strain in stress–strain curves (Fig. 2a–f). The hydrogel without CNFs showed ordinary tensile strength (0.82 MPa) at a high fracture elongation (740%) and compressive performance (4.11 MPa) at 90% strain. The mechanical properties of the gel system were substantially improved by the addition of CNFs, achieving the tensile stress of 2.01 MPa, elongation at break of 992%, and compression strength of 15.89 MPa for the PVA/CNF-4/TA/Gly organohydrogel (Fig. 2a–b). The Young’s modulus of the PVA/CNF-4/TA/Gly organohydrogel increased from 0.16 to 0.63 MPa, reflecting an increase of 3.94 times; and the toughness increased from 2.88 to 10.41 MJ/m³, indicating an increase of 3.61 times, compared to those of the hydrogels without CNFs (Fig. 2c). A further increase in CNF content to 5% leads to a decrease in the mechanical properties of the hydrogel (Fig. 2a–c). CNF is widely known to agglomerate more crystallites at higher contents, resulting in a non-uniform distribution in the hydrogel. The PVA/CNF/TA/Gly organohydrogel exhibited excellent mechanical properties because of the formation of a large number of strong and complex cross-linked networks and the strengthening effects of the CNFs.

After the optimal CNF content was determined, NaCl was applied to the hydrogel as an electrolyte, resulting in an ionic conducting hydrogel. As shown in Fig. 2d-f, the mechanical properties of the hydrogel decreased after the addition of NaCl compared to the PVA/CNF-4/TA/Gly organohydrogel. These reductions were primarily attributed to the addition of sodium salts to the gel, breaking the three-dimensional network structure of the gel, which affected the stability and mechanical properties of the gel. Similarly, the disruption of the gel network structure by NaCl can be confirmed by the higher solvent loss in the hydrogel containing NaCl, implying that the NaCl disrupted part of the hydrogen bonds was formed by the glycerol and water molecules (Fig. 3c–d). Although the flexibility and mechanical properties were affected, the organohydrogel still achieved excellent mechanical properties—that is, 1.31 MPa stress and 740% elongation at 1% NaCl content (Fig. 2d). Compared with the PVA/CNF/TA/Gly/NaCl-1 organohydrogel, the PVA/CNF/TA/Gly/NaCl-2 organohydrogel exhibited higher tensile stress (1.38 MPa), elongation at break (869%), compressive stress (13.85 MPa), Young’s modulus (0.56 MPa), and toughness (6.60 MJ/m³) (Fig. 2d-f). The increase in mechanical properties is due to the salting-out effect of NaCl leading to chain entanglement. With the
addition of high concentrations of NaCl to the hydrogel, the mechanical properties of PVA/CNF/TA/Gly/NaCl-5 organohydrogel significantly decreased, the tensile stress, elongation at break, compressive stress, Young’s modulus, and toughness decreased to 0.60 MPa, 683%, 6.45 MPa, 0.13 MPa, and 2.06 MJ/m³, respectively (Fig. 2d-f). The aforementioned mechanical property results showed that the salting-out effect of the low NaCl concentration in hydrogels promoted chain entanglement, improved the mechanical properties of hydrogels. By contrast, the high NaCl concentration (over 2%) led to excessive chain entanglement or microcrystalline zones in the polymer thus adversely affecting the hydrogel mechanical properties (Peng et al. 2018). The effect of tannic acid on the tensile properties of the hydrogels was also tested. As the TA content increased from 0 to 0.8 g, the elongation at break of the organohydrogel
increased from 420 to 869%, reflecting a 2.07 x increase, indicating that the introduction of TA effectively improved the tensile properties of the organohydrogel (Fig. S1). Moreover, a PVA/CNF/TA/Gly/NaCl-2 organohydrogel with a thickness of 1.5 mm and a width of 4 mm can easily lift 2 kg of mass without breaking, and the elongation at break was 869%, proving its excellent mechanical properties and excellent stretchability (Fig. 2h-i).

The FTIR characterization of the hydrogel was also conducted to further analyze the influence of different components on the performance of the hydrogel. The FTIR spectra revealed that the PVA/CNF hydrogel induced hydrogen bond stretching vibration at 3285 cm⁻¹ and C-O at 1035 cm⁻¹ (Fig. 2g). Owing to the formation of hydrogen bonds, the two characteristic stretching bonds of the PVA/CNF/TA hydrogel moved to 3281 and 1034 cm⁻¹. The effect of hydrogen bonds between glycerin and PVA was stronger; thus, the characteristic stretching bands of the PVA/CNF/TA/Gly organohydrogel continued to move toward the lower wavenumber (3273 cm⁻¹).

For the PVA/CNF/TA/Gly/NaCl organohydrogel, the solvation effect of salt ions weakened the effect between glycerin and water, and an increase in free glycerin caused the characteristic stretching bands to move to 3289 cm⁻¹. The addition of sodium chloride can destroy the polymer network structure and the hydrogen bond structure of the gel network as the effect of salting-out (Tao et al. 2017; Briscoe et al. 2000). The disruption of the network structure of the polymer can also be verified by the decrease in the mechanical properties of the organohydrogel after the addition of NaCl (Fig. 2d-f). Although NaCl lost part of the mechanical properties, the PVA/CNF-4/TA/Gly/NaCl-2 organohydrogel still exhibited good mechanical properties for the application of flexible wearable devices. Therefore, PVA/CNF-4/TA/Gly/NaCl-2 organohydrogel was selected for further assembly of flexible devices and other performance tests in the following section.
Environment tolerance of the organohydrogel

Water loss has been one of the challenges to the application of hydrogels. Conventional hydrogels contain large amounts of unbound water that can be easily lost in harsh environments. This characteristic causes hydrogels to wrinkle and harden and lose their original properties, thereby restricting their service life. Therefore, the realization of hydrogel sensors with water retention is of practical significance. As shown in Fig. 3a, conventional hydrogels freeze at -50 °C (cannot be pressed) and dry and harden at 60 °C. By contrast, the introduction of glycerol into the hydrogel successfully limits the evaporation of water in hot environments and the formation of ice crystals from low-temperature water molecules. As shown in Fig. 3b, the organohydrogels did not freeze at -50 °C and their mass remained almost constant (mass changed from the initial 4.7 g to 4.2 g), while the sample mass changed from 5.0 g to 2.7 g after
24 h in the oven at 60 °C. In contrast, the hydrogels dried and hardened after 24 h in the oven (mass changed from 3.3 to 0.8 g). Solvent loss is the measure of the water retention performance of hydrogels in various environments and is determined by \((W_t - W_0)/W_0\), where \(W_t\) and \(W_0\) represent the instantaneous weight and initial hydrogel weight, respectively (Chen et al. 2018). Figure 3c shows that, on Day 8, the solvent loss of the PVA/CNF/TA hydrogel at 25 °C (relative humidity of 40%) is constant and that 69% of the solvent is lost. This observation suggests that after 8 d, the glycerin-free gel almost completely lost its moisture. The hydrogels containing glycerin lost only 18% of their solvent in 8 d, effectively locking in moisture attributed to the formation of strong hydrogen bonds between glycerin and water. The solvent loss rate increased to 43% when sodium salt was added to the gel system. This was due to the fact that the salting effect of salt ions weakened the interaction between glycerol and water (Fig. 2g), increasing the amount of unbound water in the gel and leading to an increase in the solvent loss rate at room temperature for the organohydrogels containing NaCl. The results were consistent with the decrease in the mechanical properties of the sodium salt organohydrogel (Fig. 2d–e). Solvent loss increased in the extreme environment at 60 °C and was faster than that at room temperature. Solvent loss stabilized at 580 min, with
the PVA/CNF/TA, PVA/CNF/TA/Gly/NaCl, and PVA/CNF/TA/Gly hydrogels losing by 72%, 47%, and 30% of the original hydrogel weight, respectively (Fig. 3d). The reason was that the unbound and weakly bound water molecules in the gel evaporated more readily at 60°C, whereas the organohydrogel retained its stability and mechanical properties. As shown in Fig. 3e, the DSC test evaluated the effect of glycerol on the freezing resistance of the hydrogels. For the PVA/CNF/TA/NaCl hydrogel, the observed peak at -6.5°C can be considered as free water icing inside the hydrogel. When glycerol was introduced, no ice crystal peaks were observed within the temperature range of the DSC testing, indicating that the organohydrogel hardly froze within the testing range. These findings suggest that the organohydrogel can be used in flexible wearable devices even under extreme conditions.

Conductivity of the organohydrogel

The organohydrogel exhibited good ionic conductivity because of the presence of Na⁺ and Cl⁻ and the contribution of the glycerol–water binary solvent. The variation in relative resistance at different strains (from 20 to 100%) is presented in Fig. 4a, showing the strain sensitivity of the organohydrogel as a strain sensor. The gauge factor (GF, where a large value indicates high sensitivity) was further calculated to assess the strain sensitivity of the organohydrogel. GF was derived from the slope of the change in relative resistance (R - R₀/R₀) versus strain (Fig. 4b) (Dang et al. 2019; Chen et al. 2019a). As shown in Fig. 4b, three linear response regions can be obtained by linearly fitting the equations, where the relative resistance of the hydrogel increased linearly at 0–150% strain with a GF of 1.98 (R² = 0.989), and further increased the strain to 300% showing a GF of 4.84 (R² = 0.987). When the strain increased to 500%, the GF reached a maximum value of 8.54 (R² = 0.998). The high GF in the range of measured tensile strains indicated that the PVA/CNF/TA/Gly/NaCl-2 organohydrogel exhibited superior sensitivity to strain, accompanied by a wide sensing range (Chen et al. 2020). Therefore, the stability of the organohydrogel in an open environment was investigated to better assess their stability in the environment. Although the conductivity of the PVA/CNF/TA/Gly/NaCl-2 organohydrogel gradually decreased with time, the conductivity of this sample after 7 d remained high (0.75 S/m) (Fig. 4d). The durability was tested by performing 1000 tensile tests at 50% strain and showed a stable resistance signal, which was conducive to the stability and reusability of the sensor in real life (Fig. 4e).

Wearable flexible strain sensors in human motion monitoring

With its excellent mechanical properties, good strain sensitivity, and signal stability considered, the organohydrogel was assembled into a tensile strain sensor to detect human electrical signals. The strain sensors were applied to different joints of the human body to check the complex human motion in real-time.
The change in relative resistance \((R - R_0)/R_0\) under different bending angles of the finger increased rapidly when the finger was bent at 90° (Fig. 5a) and returned to the original value when the angle was 0° (Online Resource 1). Notably, after repeated cycles, the change in relative resistance still maintained similar peaks and valleys, indicating the good stability, immediacy, and accuracy of the organohydrogel as a strain sensor in detecting human signals. The motion of similar wrist and elbow joints can also be accurately monitored via assembly into a strain sensor (Fig. 5b–c). The PVA/CNF/TA/Gly/NaCl-2 organohydrogel strain sensors can also detect subtle movements. The organohydrogel was applied to the throat of the volunteer to detect sound vibrations. When the words “Hi,” “I,” and “Hello,” were spoken, the signal could be detected, with different resistance peaks and valleys (Fig. 5d–f). This finding suggests that as a strain sensor, the organohydrogel exhibited superior sound recognition capability. The ability of the PVA/CNF/TA/Gly/NaCl-2 organohydrogel to detect electrical signals under extreme conditions was also evaluated to further demonstrate its environmental tolerance. Notably, the organohydrogel sensor exhibited good strain sensing capability even after storage for 12 h at -50 °C and 60 °C and after storage for 30 d in an open environment (Online Resource 2). The organohydrogel sensor still showed satisfactory reproducible and stable resistance signals during finger bending (Fig. 5g–i). Therefore, the PVA/CNF/TA/Gly/NaCl-2 organohydrogel-based sensor can stably sense human motion signals with different motion amplitudes for a long time even in extremely harsh environments, proving its great potential for application in wearable devices.

Conclusions

In summary, a novel flexible PVA/CNF/TA/Gly/NaCl organohydrogel with high tensile properties, anti-freezing and anti-drying features, long-term stability at room temperature, and high strain sensitivity was prepared using a simple one-pot synthesis strategy. The formation of strong hydrogen bonds between the water and glycerol molecules introduced into the hydrogel inhibited the formation of hydrogel ice crystals even at low temperatures (e.g., −50 °C) and prevented the evaporation of water at high temperatures (e.g., 60 °C). Moreover, good performance was observed even after 30-d storage. The introduction of NaCl conferred good ionic conductivity \((\sim 0.86 \text{ S/m})\) and high strain sensitivity \((\text{GF} = 8.54)\) on the organohydrogel. These characteristics allow the use of the organohydrogel as a wearable electronic device that can detect and differentiate human motion, such as multiple-strain movement (joint movement) and subtle movement (voice in the throat). It is believed that this research provides a strategy for the potential applications of CH flexible wearable devices and human health monitoring in daily life and extreme environments.

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Conflict of interest

The authors declare that they have no conflict of interest.

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