



Dual-salt-induced polyionic hydrogels with controlled swelling for self-healable underwater strain sensors

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ABSTRACT

Although ionic hydrogels are promising soft conductors for underwater strain sensing and human-machine interfaces, prolonged exposure to aqueous or saline environments can cause these hydrogels to exhibit swelling, ion leakage, conductivity drift, and loss of sensing repeatability. In this study, we present a dual-salt hydrogel referred to as PGNCM, which is composed of poly(vinyl alcohol) (PVA), glycerol, NaCl, CaCl₂, and carboxyl-functionalized multiwalled carbon nanotubes (fMWCNTs), for underwater sensing. NaCl is associated with salting-out-assisted PVA chain ordering, whereas CaCl₂ introduces divalent ion-mediated interactions within the PVA/fMWCNT network. This dual-salt environment reduces swelling while preserving mechanical robustness, electrical conductivity, and self-healing. PGNCM exhibited a swelling ratio of ~54 % after 48 h in artificial seawater, retaining 87 % of its initial conductivity after self-healing. As a strain sensor, it shows a linear response over 0–200 % strain ($GF = 1.27$, $R^2 = 0.997$) and retains a cyclic resistance response over 1000 cycles at 100 % strain. The sensing response is retained even after two days of immersion in deionized water and seawater, and it remains stable over 5–40 °C. A single-sensor element enables underwater Morse code communication and robotic-fish locomotion tracking under direct aqueous exposure, demonstrating the potential of the dual-salt freeze-thaw strategy for hydrogel-based underwater sensing applications.

1. Introduction

Hydrogels are three-dimensional crosslinked polymer networks that can absorb large amounts of water while retaining structural integrity [1–3]. Hydrogel-based materials exhibit advantageous properties such as tunable ionic and electronic conductivities, high water content, and mechanical compliance [4–8]. These properties have enabled advancements in energy storage [9], wearable electronics [10], human-machine interfaces [11], flexible electronics [12], and soft robotics [13]. Recent multifunctional conductive gel sensors have further demonstrated integrated capabilities in motion monitoring, multimodal sensing, and human-machine interfaces [14–17]. Conductive hydrogels can be broadly designed using ion-conducting components, electronically conductive fillers or polymers, or hybrid ionic-electronic networks [18]. Among these, ionic hydrogels are particularly attractive for soft strain sensing because charges are carried by mobile ions distributed throughout the water-swollen network, which causes the conductive medium to remain soft and deform with the matrix.

Ionic hydrogels are typically prepared by incorporating ionic

components into a hydrogel matrix to impart ionic conductivity to an otherwise insulating network. Common ionic components include inorganic salts [5], ionic liquids [19], acids [20], and polyelectrolytes [21]; among these, inorganic salts are especially convenient because they not only supply charge carriers but also modify the polymer network. Through Hofmeister-type salting-in and salting-out interactions [22], salt ions can either reinforce or disrupt polymer-chain aggregation, crystallinity, and hydrogen bonding, tuning the mechanical properties of the gel [23]. Wang et al. reported that salt ions can diffuse into the semicrystalline and crystalline regions of polymer networks and weaken interchain hydrogen bonding, thereby contributing to the frequently observed trade-off between ionic conductivity and mechanical strength [24]. Although single-salt ionic hydrogels are simple and effective, they often couple conductivity with mechanical properties, complicating independent optimization. A further limitation arises underwater: when a single-salt ionic hydrogel is immersed, the osmotic and concentration gradients between the gel and surrounding medium can draw water into the network and drive ions out. This can lead to swelling, ion leakage, and conductivity drift that degrade

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long-term sensing stability [25]. Addressing these coupled challenges requires strategies for independently regulating ion transport, network mechanics, and environmental stability.

Compared to single-salt systems, dual-salt designs provide an additional degree of freedom for regulating ion–ion and ion–polymer interactions. To date, such approaches have been more commonly explored in battery-related electrolyte systems where pairing two salts enhances ionic conductivity and electrochemical stability [26–36]. The principle underlying this enhancement is that chemically distinct ions play complementary roles within a single network. This principle is equally attractive for sensing hydrogels because it offers a route to simultaneously tune the ionic conductivity, mechanical robustness, and environmental stability instead of trading one against another. Studies have attempted to realize underwater stability by suppressing swelling: both by increasing the network constraint, which improves the elastic restoring pressure of the gel [37], and by modifying the polymer–water and polymer–polymer interactions through additional physical or chemical crosslinking [38–42]. Despite this potential, dual-salt strategies remain largely unexplored in poly(vinyl alcohol) (PVA) hydrogels for underwater conductive sensing, wherein dimensional stability, conductivity, and mechanical integrity must be satisfied simultaneously.

In this study, we report a polyionic, conductive, and self-healing hydrogel referred to as PGNCM. The proposed PGNCM hydrogel is composed of PVA, glycerol, a dual-salt combination of sodium chloride (NaCl) and calcium chloride (CaCl_2), and carboxyl-functionalized multiwalled carbon nanotubes (fMWCNTs). The hydrogel is assembled through physical crosslinking using a one-pot heat-and-stir process followed by two freeze–thaw cycles. The dual-salt pair is selected to address the aforementioned gap in underwater dimensional stability. NaCl and CaCl_2 are selected to introduce complementary ion-mediated effects within a single physical network, where NaCl is associated with network stabilization through salting-out-assisted PVA-chain ordering, whereas CaCl_2 introduces divalent ion-mediated interactions within the PVA/fMWCNT network. Na^+ is abundant and Ca^{2+} is present as a major divalent cation in seawater, thereby, this composition is expected to reduce the osmotic driving force for swelling and salt washout, thereby improving the dimensional stability of the gel during saline immersion. The resulting hydrogel exhibits high mechanical strength, toughness, and recoverable conductive-network integrity, represents a swelling ratio of $\sim 54\%$ after 48 h in artificial seawater, and retains high conductivity and sensing performance. As a strain sensor, it provides a linear and durable response during underwater operation. Further, we

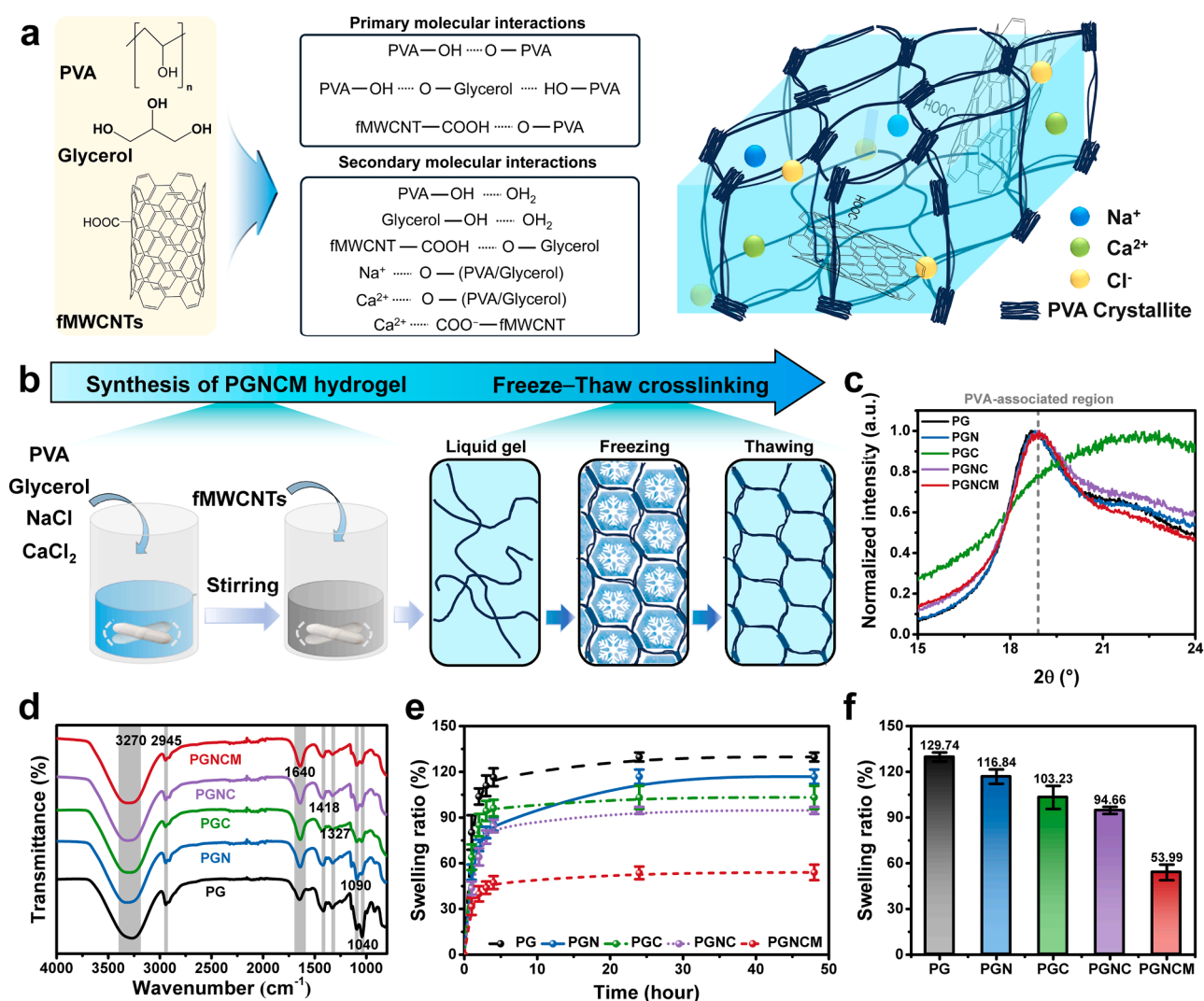


Fig. 1. Fabrication and structural characterization of the PGNCM hydrogel. (a) Schematic of the molecular network and principal intermolecular interactions within the PGNCM matrix. (b) Synthesis workflow highlighting the freeze–thaw-driven physical crosslinking step. (c) Enlarged XRD patterns of PG, PGN, PGC, PGNC, and PGNCM hydrogels in the PVA-associated region ($15\text{--}24^\circ$). (d) FTIR spectra of PG, PGN, PGC, PGNC, and PGNCM hydrogels showing the chemical composition and FTIR spectral changes associated with the introduction of NaCl, CaCl_2 , and fMWCNTs. (e) Swelling kinetics of the five hydrogels in artificial seawater over 48 h. (f) Swelling ratios after 48 h showing the reduced swelling of PGNCM under saline conditions.

demonstrate two underwater functions under saline conditions using a single-sensor element. These results highlight the ability of the dual-salt network to maintain dimensional stability, conductivity, and sensing reliability during underwater operation.

2. Results and discussion

2.1. Concept of the PGNCM hydrogel

The PGNCM hydrogel was designed as a fully physical dual-salt-mediated network in which each component plays a distinct role (Fig. 1a). PVA provides a semicrystalline backbone with hydroxyl-rich chains supporting both freeze–thaw-induced crystallite formation and extensive hydrogen bonding. Glycerol acts as a hygroscopic plasticizer and co-solvent, suppressing macroscopic ice growth during freezing and preserving chain mobility throughout the network. Unlike post-immersion solvent replacement used in some glycerol organo-hydrogels [43], glycerol is incorporated in situ to avoid an additional solvent-exchange step. NaCl was introduced to assist PVA-chain aggregation and ordering through salting-out during freeze–thaw processing, consistent with previous reports on ion-regulated PVA networks [44]. In parallel, CaCl_2 provides divalent ions capable of interacting with oxygen-containing groups in the PVA/fMWCNT network. Ca^{2+} -mediated interactions with hydroxyl and carboxylate groups have been reported in related systems [45]. In the present hydrogel, such interactions are considered a plausible dynamic contribution rather than a uniquely resolved coordination state. High-resolution X-ray photoelectron spectroscopy (XPS) analysis further confirmed the presence of Ca in the CaCl_2 -containing formulations, whereas no discernible Ca 2p signal was observed for the CaCl_2 -free PVA/glycerol/NaCl/fMWCNT (PGNM) sample (Fig. S5). However, the nearly identical Ca $2p_{3/2}$ binding energies of the CaCl_2 -containing formulations did not resolve a distinct carboxylation-dependent Ca^{2+} coordination state. The fMWCNTs serve primarily as reinforcing nanofillers, with interfacial hydrogen bonding between their carboxyl groups and PVA hydroxyl groups, and provide oxygen-containing interfacial sites that may participate in Ca^{2+} -mediated interactions, while contributing secondarily to bulk electrical transport.

Following this rationale, the PGNCM hydrogel was synthesized through a one-pot heat-and-stir step followed by two freeze–thaw cycles (Fig. 1b). NaCl and CaCl_2 were initially co-dissolved in deionized water (DIW) to form a homogeneous dual-salt precursor. Subsequently, PVA and glycerol were added and dissolved via stirring at an elevated temperature to yield a transparent viscous solution. A pre-dispersed fMWCNT suspension was subsequently introduced and stirred for 1 h to ensure the uniform distribution of the conductive filler. The resulting mixture was poured into molds and subjected to two freeze–thaw cycles, where each cycle comprised freezing at -18°C for 24 h and thawing at room temperature for 1 h. This process converted the viscous precursor to a self-standing mechanically robust hydrogel. Four samples, namely, PVA/glycerol (denoted as PG), PVA/glycerol/NaCl (PGN), PVA/glycerol/ CaCl_2 (PGC), and PVA/glycerol/NaCl/ CaCl_2 (PGNC), were prepared using the same procedure with the selective omission of NaCl, CaCl_2 , and fMWCNTs, thereby enabling each contribution to be assessed in subsequent characterizations.

To examine composition-dependent changes in the PVA-associated structure, X-ray diffraction (XRD) analysis was performed on the freeze-dried hydrogels (Fig. 1c). In the enlarged $15\text{--}24^\circ$ region, the NaCl-containing PGN, PGNC, and PGNCM formulations retained a distinct diffraction maximum near 19° , whereas the CaCl_2 -only PGC exhibited a broader profile. These differences are consistent with the contrasting effects of salting-out and salting-in ions on PVA-chain aggregation reported previously, and support the proposed role of NaCl in assisting PVA-chain ordering without constituting direct evidence of Na^+ -induced crystallization. The full-range patterns further showed sharp reflections consistent with crystalline NaCl in the NaCl-containing

formulations (Fig. S6).

Chemical interactions underlying the dual-salt-mediated network were probed by Fourier-transform infrared (FTIR) spectroscopy of the as-prepared hydrogels (Fig. 1d). All formulations displayed the characteristic absorption bands of the plasticized PVA matrix (Fig. S1). A broad O–H stretching envelope at $3200\text{--}3500\text{ cm}^{-1}$ originated from PVA, glycerol, and hydrogen-bonded water. The C–H stretching band of the methylene backbone appeared at 2945 cm^{-1} , and CH_2 -related bending modes were observed near 1418 cm^{-1} . Overlapping C–O stretching modes at 1090 and 1040 cm^{-1} were within the PVA C–O fingerprint region [46,47]. The O–H stretching maximum of the salt-free PVA/glycerol/fMWCNT (PGM) control ($\sim 3270\text{ cm}^{-1}$) remained close to that of PG ($\sim 3272\text{ cm}^{-1}$), whereas the salt-containing formulations exhibited O–H maxima at $3295\text{--}3315\text{ cm}^{-1}$ (Table S1). These spectral changes were consistent with salt-associated perturbations of the hydrogen bonding [48,49].

Subsequently, the dimensional stabilities of the five hydrogels were evaluated by tracking their mass uptake in artificial seawater over 48 h (Fig. 1e). All formulations exhibited two-stage swelling profiles. The first stage involved rapid uptake within the first 6 h, which was dominated by the capillary and surface diffusion of saline water into the as-prepared network. This was followed by a slower change in mass over 24–48 h. The five formulations exhibited distinct swelling behaviors, differing in both the extent of uptake and rate of early-stage swelling. PG, which lacked any salt-mediated reinforcement, swelled most extensively and showed the steepest initial slope, reflecting a loosely crosslinked network in which only freeze–thaw-induced PVA crystallites and hydrogen bonding resisted solvent ingress. PGN exhibited a markedly suppressed uptake relative to PG, which was attributed to the salting-out-assisted chain ordering of the PVA backbone [23,50]. This reduced the free volume available to the incoming water molecules and increased the elastic restoring pressure of the network. PGNCM exhibited the lowest uptake throughout the 48 h window. This behavior is consistent with the combined influence of the dual-salt environment and fMWCNT incorporation on the physical network and water uptake of the hydrogel.

The swelling ratios of PG, PGN, PGC, PGNC, and PGNCM after 48 h were 129.74, 116.84, 103.23, 94.66, and 53.99 %, respectively, as depicted in Fig. 1f. The decrease in the ratio from PG to PGNCM upon the sequential introduction of NaCl, CaCl_2 , and fMWCNTs was consistent with the cumulative contribution of these components. The 53.99 % swelling ratio of PGNCM after 48 h was comparable to values reported for PVA-based hydrogels developed for underwater sensing. For example, a recent PVA-based hydrogel combining freeze–thaw processing and metal-ion crosslinking exhibited an equilibrium swelling ratio of approximately 50 % in water [51], while a salting-out PVA/MXene hydrogel swelled by 75 % in water [52], whereas an amylopectin/NaCl PVA hydrogel swelled by only 3.55 % in seawater [53]. In comparison, conventional freeze–thaw PVA hydrogels are generally water-rich networks whose swelling capacity and water uptake depend strongly on the gelation and crosslinking conditions [54, 55]. Consistent with this general behavior, the PG control in this study exhibited a substantially higher swelling ratio (129.74 %) than that of PGNCM (53.99 %). Even lower swelling ratios have been reported for systems that incorporate zwitterionic comonomers or additional covalent crosslinking, including covalently polymerized zwitterionic networks ($\sim 9\%$ in water) [56], covalently crosslinked thermoresponsive nonswelling networks [57], and Hofmeister-assisted zwitterionic PVA networks that essentially show no net swelling in water and seawater [58]. However, these approaches typically introduce additional copolymer synthesis steps or rely on chemical crosslinkers with associated biocompatibility concerns [54]. PGNCM attains this saline dimensional stability without zwitterionic comonomers, covalent crosslinkers, or hydrophobic encapsulation, while retaining electrical conductivity and self-healing. Moreover, the NaCl/ CaCl_2 pair contains Na^+ , which is an abundant seawater cation, and Ca^{2+} , which is a major divalent cation

present in seawater. This compositional similarity can reduce ion-exchange-driven swelling and salt washout under saline conditions. The role of glycerol in the environmental stability of PGNCM was further evaluated by differential scanning calorimetry (DSC) and weight-retention measurements. Compared with the glycerol-free PNCM hydrogel (PGNCM formulation without glycerol), PGNCM exhibited lower crystallization and melting peak temperatures and a lower melting enthalpy, consistent with suppressed ice crystallization and a reduced amount of freezable water (Fig. S7 and Table S3). In addition, PGNCM retained 32.16 ± 0.38 % of its initial weight after 7 days, compared with 19.46 ± 0.41 % for PNCM (Fig. S8), demonstrating improved moisture retention under the tested storage conditions. Similar polyol-mediated improvements in anti-freezing and moisture-retention behavior have been reported in multifunctional gel sensors [15,59–61]. The dimensional stability established in this section provides the structural basis for the mechanical, viscoelastic, and strain-sensing characterization presented in Sections 2.2–2.4.

2.2. Mechanical properties

The dual-salt design rationale established in Section 2.1 suggests a characteristic mechanical response. The NaCl-containing formulations are expected to retain a comparatively ordered PVA framework, whereas CaCl_2 and fMWCNT incorporation introduce additional ion-mediated and polymer–filler interactions that may affect extensibility and

energy dissipation. The uniaxial tensile testing of PGN, PGC, PGNC, and PGNCM hydrogels was conducted to verify this prediction (Fig. 2a–c). Among the PGN, PGC, and PGNC hydrogels, the small-strain stiffness decreased in the order PGNC, PGN, and PGC, with Young's moduli of 177, 171, and 119 kPa, respectively (Fig. 2b). Both PGN and PGNC contained NaCl and exhibited a similar stiffness range, consistent with the NaCl-associated PVA ordering discussed in Section 2.1, whereas the CaCl_2 -only PGC hydrogel was markedly more compliant. The reported salting-in tendency of CaCl_2 in freeze–thaw PVA network [23] could attenuate the freeze–thaw-induced chain ordering, thereby contributing to the lower modulus. Further, the PGN hydrogel exhibited the highest ultimate tensile strength (UTS) among salt-only samples (1195 kPa). However, it had the lowest strain at break (303 %), indicating a comparatively less extensible response than the CaCl_2 -containing PGC and PGNC hydrogels. Both PGC and PGNC hydrogels failed at considerably larger strains (~410 %), indicating enhanced extensibility relative to that of the PGN hydrogel, yielding UTS values of 1037 and 1160 kPa, respectively. Compared to the PGNC hydrogel, PGNCM exhibited a slightly lower modulus and strain at break, while exhibiting a higher UTS of 1256 kPa. The four toughness values calculated from the integrated area under the stress–strain curves did not follow the UTS trend. The PGNC hydrogel exhibited the highest toughness (1889 kJ/m^3) followed by the PGNCM (1769 kJ/m^3), PGC (1625 kJ/m^3), and PGN (1495 kJ/m^3) hydrogels (Fig. 2c). The PGNCM hydrogel combined the highest UTS in the series with a toughness comparable to that of the PGNC

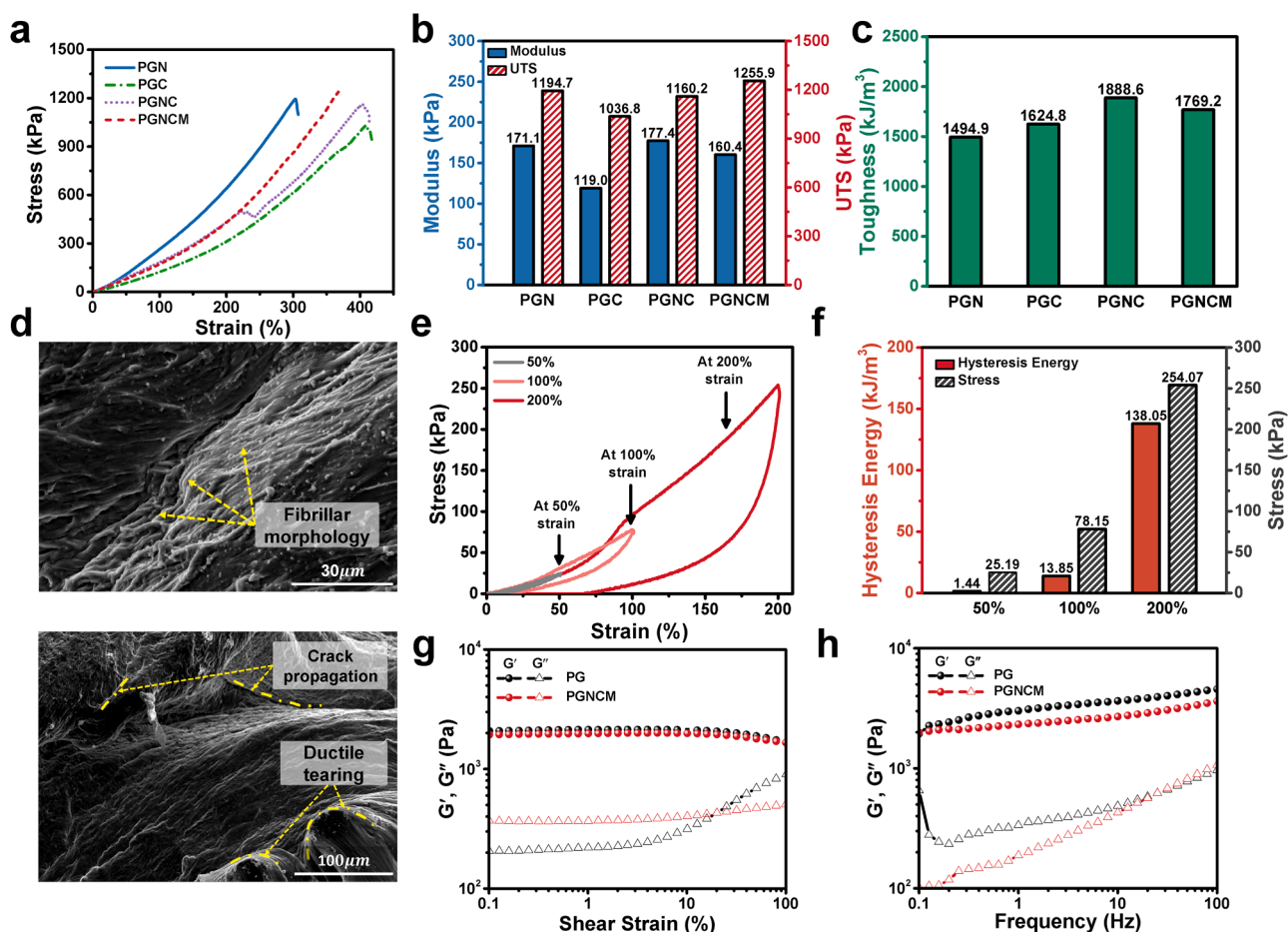


Fig. 2. Mechanical and rheological characterizations of the PGNCM hydrogels. (a) Representative tensile stress–strain curves of the PGN, PGC, PGNC, and PGNCM hydrogels. (b) Young's modulus, UTS, and (c) toughness of the PGN, PGC, PGNC, and PGNCM hydrogels derived from the curves in (a). (d) SEM images of the PGNCM hydrogel: bulk-surface views of fibrillar PVA morphology and tensile fracture-surface views of crack-deflection and ductile-tearing features. (e) Loading–unloading cycles of the PGNCM hydrogel at strain amplitudes of 50, 100, and 200 % (first cycle shown; five-cycle data at each amplitude in Fig. S2). (f) Hysteresis energy and peak stress of the first loading–unloading cycle extracted from (e). Rheological response of PG and PGNCM: (g) strain sweep and (h) frequency sweep showing the storage and loss moduli (G' and G'' , respectively).

hydrogel. The stretching rate-dependent tensile behavior of PGNCM was additionally evaluated at stretching rates of 50, 100, and 200 mm/min (Fig. S9). The UTS increased from 0.70 ± 0.09 to 1.28 ± 0.11 MPa and the toughness increased from 0.87 ± 0.12 to 1.54 ± 0.22 MJ/m³ as the stretching rate increased from 50 to 200 mm/min, whereas the elongation at break remained nearly unchanged at ~ 365 %. This behavior was consistent with the rate-dependent viscoelastic response of the physically crosslinked PGNCM network.

Scanning electron microscopy (SEM) was conducted to characterize the network structure of the PGNCM hydrogel before and after tensile failure (Fig. 2d). The bulk surface comprised densely packed fibrillar bundles (widths in the range of 1–3 μ m) that lay roughly parallel to one another over the field of view, matching the fibrillar PVA-rich scaffold described in Section 2.1. In contrast, the tensile fracture surface was dominated by features of large-scale plastic deformation. Both the deflected, multitiered crack path and pervasive ductile-tearing texture have been associated with enlarged crack-tip dissipative zones in tough hydrogels [62], and their coexistence is consistent with the energy-dissipating failure mode that the cyclic-loading hysteresis probes mechanically (Fig. 2e and f).

The energy-dissipating behavior of the PGNCM network was probed by cyclic loading–unloading at strain amplitudes of 50, 100, and 200 % (Fig. 2e). The three cycles enclosed hysteresis loops with markedly different magnitudes. At 50 % strain, the loop was nearly closed (dissipated energy 1.44 kJ/m³ and peak stress 25.19 kPa). A discernible hysteresis loop appeared at 100 % strain (13.85 kJ/m³ and 78.15 kPa), and the loop expanded substantially at 200 % strain (138.05 kJ/m³ and 254.07 kPa) (Fig. 2f). The dissipated fraction of the loading work increased from ~ 1 % at 50 % strain to 7 and 44 % at 100 and 200 % strain, respectively. This trend was consistent with progressively greater rearrangement of reversible physical interactions within the PGNCM network at larger strains [63]. This threshold separated the low-damage regime from the high-dissipation regime. This was relevant for repeated strain sensing and toughness enhancement. The hysteresis values reported above were extracted from the first loading–unloading cycle at each amplitude. Under repeated cycling (Fig. S2), the 50 % response overlapped almost completely over the five successive cycles, whereas the 100 and 200 % responses exhibited Mullins-like softening between the first and second cycles [64]. The peak stress decreased from 78 to 73 kPa at 100 % strain and from 254 to 238 kPa at 200 % strain, after which the second-to-fifth cycles overlapped closely. This behavior indicated a partial disruption or rearrangement of the dynamic junctions during the first large-deformation cycle. Thereafter, the network anchored by the intact freeze–thaw-induced PVA crystalline framework delivered a reproducible steady-state response that supported continued large-deformation cycling without progressive damage.

Small-amplitude oscillatory shear rheology was used to probe the same network from a linear-viscoelastic perspective. In the strain sweep (Fig. 2g, $\omega = 10$ rad/s), both PG and PGNCM hydrogels displayed a wide linear-viscoelastic plateau in which storage modulus G' was above loss modulus G'' , indicating a solid-like, gel-state response in both samples. The two samples reached comparable elastic plateaus ($G' = 2.10$ kPa and 1.96 kPa for PG and PGNCM, respectively). However, PGNCM exhibited a higher loss modulus ($G'' = 0.37$ kPa and 0.21 kPa for PGNCM and PG, respectively), larger loss tangent ($\tan \delta = 0.19$ and 0.10 for PGNCM and PG, respectively), and wider linear-viscoelastic window (yielding at 59 % strain and 50 % strain for PGNCM and PG, respectively). Enhancing the small-strain dissipation without raising the elastic plateau was consistent with the presence of additional dynamic interactions in PGNCM. This contributed to viscoelastic dissipation at low strain and was mechanically defined only at larger strains, where they disrupted or rearranged as sacrificial junctions (Fig. 2e and f). The frequency sweep (Fig. 2h, 0.1 % strain) showed a nearly frequency-independent $G' > G''$ over the entire 0.1–100 rad/s window, which was consistent with a physically crosslinked gel network. The loss tangent of PGNCM remained low (0.05–0.30) over the entire window. Therefore, the dual-

salt/fMWCNT modifications acted in the linear viscoelastic region by adding viscoelastic dissipation while preserving the elastic network response and extending the linear-viscoelastic window. Further, PGNCM withstood compressive loading up to 3.7 MPa at 90 % strain without macroscopic fracture (Fig. S3), indicating adequate structural robustness under compressive deformation. Under cyclic compression to 50 % strain, the peak compressive stress decreased from 28.8 kPa in the first cycle to 23.3 kPa in the second cycle and remained nearly constant through the 20th cycle (23.0 kPa), indicating a stable cyclic response after the initial stress softening (Fig. S3).

2.3. Electromechanical sensing characteristics

The bulk conductivities of the four salt-containing formulations were measured to assess the contribution of each component to charge transport (Fig. 3a). The conductivity increases from 0.24, 0.27, 0.28, and 0.30 S/m for the PGN, PGC, PGNC, and PGNCM hydrogels, respectively. To further distinguish the contribution of fMWCNTs from salt-mediated transport, the salt-free PGM control was additionally evaluated and exhibited a bulk electrical conductivity of 0.0368 S/m. In comparison, the dual-salt PGNC hydrogel without fMWCNTs already exhibited a conductivity of 0.28 S/m, close to that of PGNCM (~ 0.30 S/m). These results indicate that PGNCM is best described as a dual-salt, multi-ion ionic conduction system, in which mobile Na⁺, Ca²⁺, and Cl[−] ions serve as the principal charge-transport pathway, while fMWCNT incorporation provides only a secondary contribution under the present low loading. A similar predominance of ionic transport was reported for a related PVA/glycerol/NaCl/MWCNT hydrogel system [65]. Because the present bulk conductivity measurements do not independently resolve ionic and electronic currents, the relative contributions are assessed comparatively across the control formulations.

When subjected to uniaxial tensile strain, the sensor exhibits an increase in resistance owing to geometric deformation and strain-induced changes in the charge-transport pathways within the hydrogel network [65]. The PGNCM hydrogel sensor exhibits a near-linear $\Delta R/R_0$ response over the entire 0–200 % strain window, with gauge factor $GF = 1.27$ and coefficient of determination $R^2 = 0.997$ extracted from the linear fit (Fig. 3b). Measurements using three independently fabricated sensors yielded a mean GF of 1.267 ± 0.012 (mean $\pm$ standard deviation, $n = 3$).

The low-strain sensing capability was further evaluated under cyclic strains of 1–5 % (Fig. 3c). Distinct and repeatable resistance changes were observed even at 1 % strain, indicating a minimum detectable strain of 1 %. At larger deformations, $\Delta R/R_0$ increased systematically at strain amplitudes of 50, 100, and 200 %, with repeatable responses over successive cycles (Fig. 3d). Under a stepwise strain sequence of 20, 50, 80, 40, 10, and 20 % (Fig. 3e), the $\Delta R/R_0$ signal followed the imposed strain levels and recovered toward the baseline upon unloading.

The sensor exhibits short response and recovery times of 306 and 345 ms, respectively, at a 25 % strain step (Fig. 3f). Electrical hysteresis was further evaluated from the loading and unloading $\Delta R/R_0$ responses during a cycle to 100 % strain (Fig. 3g). The resulting loading and unloading curves showed an electrical hysteresis of ~ 5.6 %, indicating a largely reversible electromechanical response at 100 % strain.

To evaluate durability under large deformation, the sensor was subjected to 1000 continuous loading–unloading cycles at 100 % strain (Fig. 3h). The sensor maintained repeated resistance responses throughout the cycling test, while comparison of the initial and final cycles revealed a gradual baseline drift of ~ 66 %, with the unloaded $\Delta R/R_0$ baseline increasing from ~ 0.6 % to ~ 67 %. Despite this shift, the peak-to-peak response amplitude was largely preserved, changing from ~ 109 to ~ 97 % (~ 89 % retention) over the 1000 cycles. Additional stretching rate-dependent responses are provided in Fig. S10.

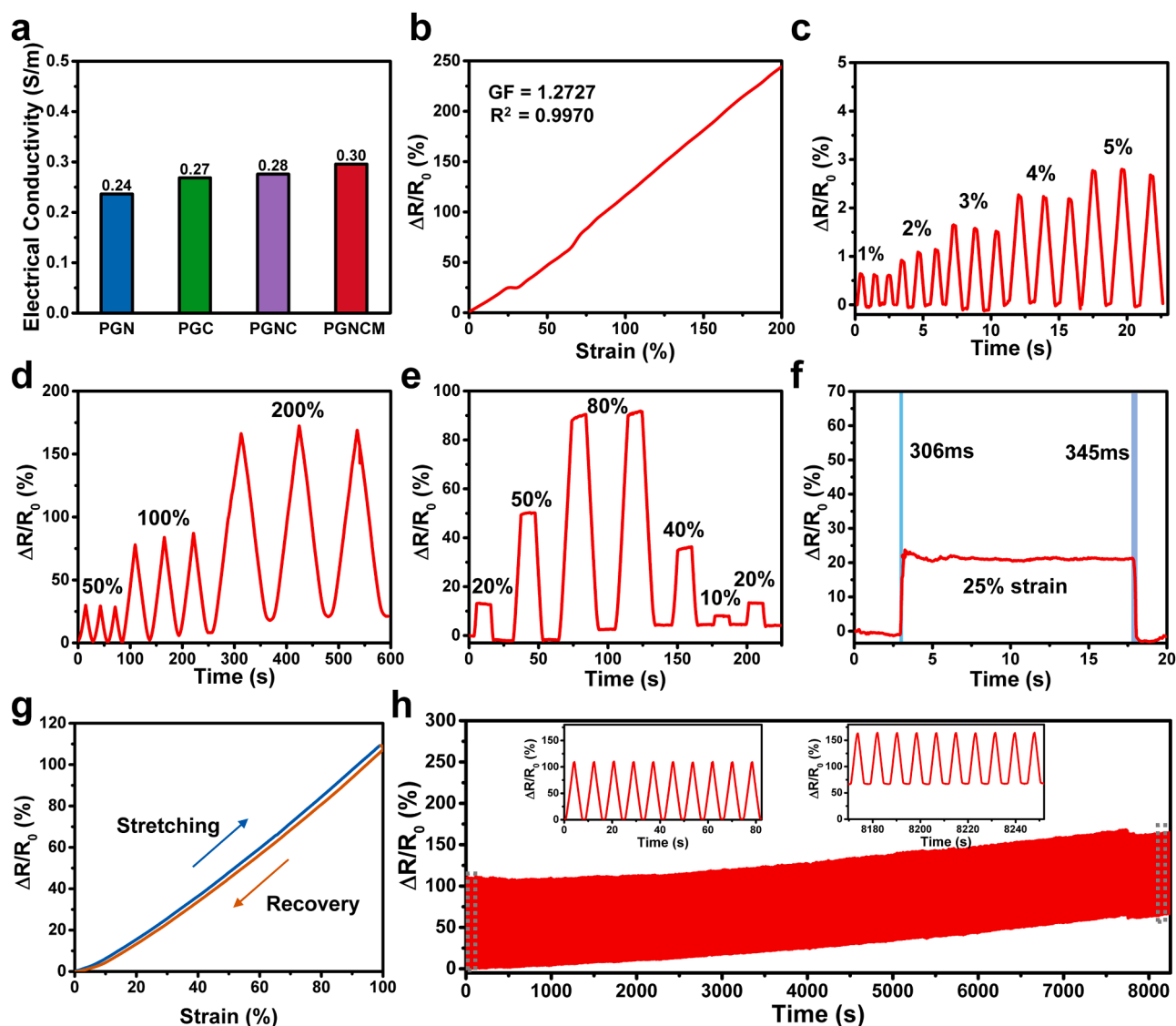


Fig. 3. Electromechanical sensing performance of the PGNCM sensor. (a) Bulk conductivities of the PGN, PGC, PGNC, and PGNCM hydrogels showing the contribution of each component to charge transport. (b) Relative resistance response ($\Delta R/R_0$) as a function of applied strain with gauge factor (GF) and linearity (R^2) extracted from the linear fit. (c) Cyclic $\Delta R/R_0$ responses at small strains of 1–5 %, demonstrating a resolvable response at 1 % strain. (d) Time-resolved $\Delta R/R_0$ responses at large strains of 50, 100, and 200 %. (e) $\Delta R/R_0$ signals under stepwise strain loading. (f) Response and recovery times at 25 % strain. (g) Loading and unloading $\Delta R/R_0$ responses during a cycle to 100 % strain, showing the electrical hysteresis of the sensor. (h) Long-term cyclic response over 1000 cycles at 100 % strain. The insets show magnified responses during the initial and final cycles.

2.4. Self-healing behavior and functional recovery

The dual-salt design rationale established in Section 2.1 suggests a potential route to healing. Glycerol, which plasticizes the network in the as-prepared state (Section 2.1), is expected to help retain polymer-chain mobility during the thawing step, thereby facilitating the reformation of reversible physical interactions across the contact zone (Fig. 1a). Accordingly, the freeze–thaw-assisted self-healing of the PGNCM hydrogel was examined first. To this end, a rectangular sample was completely severed with a razor blade, the cut faces were gently brought into contact without external pressure, and the assembly was subjected to two successive freeze–thaw (F–T) cycles, each consisting of freezing at -18°C for 24 h followed by thawing at room temperature for 1 h [54, 65] (Fig. 4a, Movie S1). Additional specimens were evaluated after 12 and 18 h of freezing during the first healing cycle to quantify the healing-duration dependence of the electrical recovery (Fig. 4c). After the second freeze–thaw cycle, the originally transected interface appeared macroscopically continuous. The healed sample withstood

manual stretching without reopening at the cut interface, indicating recovered mechanical integrity. When connected in series with a light-emitting diode (LED) circuit, it restored LED illumination, confirming electrical continuity across the healed region. During the healing freeze–thaw cycles when the cut faces were held in contact, repeated freezing and thawing promoted the reformation of the same physical interactions that formed the original gel across the cut, thereby restoring network connectivity [65,66]. This proceeds through the original physical crosslinking with no covalent or photochemical triggers; therefore, the healed hydrogel recovers substantial electrical continuity and mechanical integrity.

The proposed reintegration method is supported morphologically by the SEM imaging of the healed sample across the original cut plane (Fig. 4b). On the bulk surface (Fig. 4b, left), the original cut is no longer resolvable as a discrete feature. Fibrillar PVA bundles characteristic of the pristine PGNCM matrix (Fig. 2d) extend continuously across the field of view, with no clearly visible grooves, cleavage lines, or alignment discontinuities at this magnification. A cross-sectional view of the same

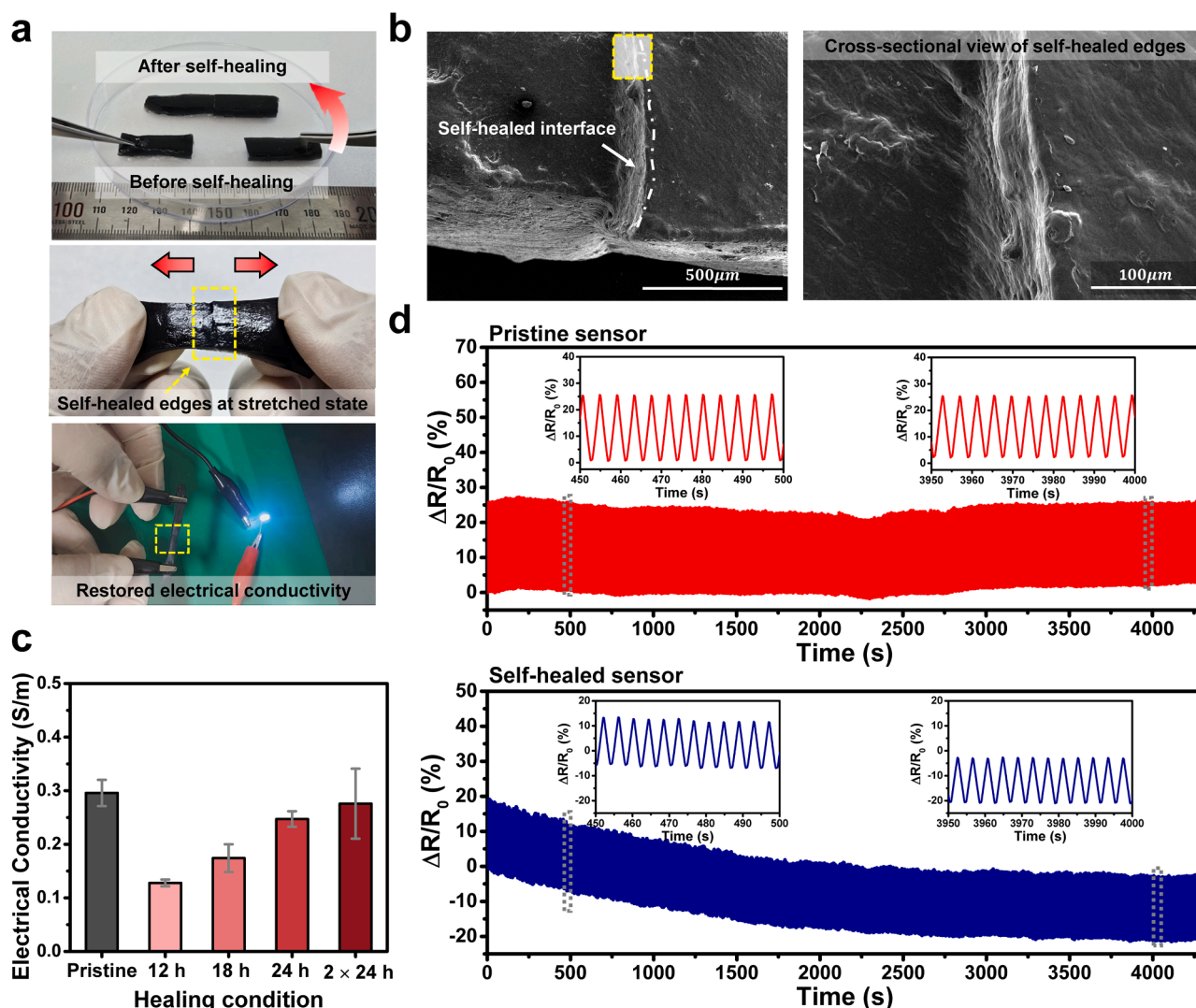


Fig. 4. Self-healing performance and functional recovery of the PGNCM hydrogel. (a) Optical images showing the macroscopic self-healing process, stretchability of the healed sample, and restoration of electrical conductivity in a circuit closed by the healed gel. (b) SEM images of the self-healed interface (left) and cross-sectional view of the healed interface (right), showing morphological continuity across the healed interface. (c) Electrical conductivity recovery after healing for 12, 18, and 24 h during the first F–T cycle and after a second 24 h F–T cycle, compared with the pristine state. (d) Long-term cyclic electromechanical responses of the pristine and self-healed PGNCM sensors over 1000 loading–unloading cycles at 25 % strain. The insets show representative responses during the early and late stages of cycling.

healed edge (Fig. 4b, right) supports this observation in the through-thickness direction. The two previously opposing faces appear continuous, with the fibrillar morphology preserved across the healed interface. Energy-dispersive X-ray spectroscopy (EDS) of the same region uniformly detects all five constituent elements (C, O, Na, Cl, and Ca) across the healed interface (Fig. S4), which is consistent with the retention of the major network constituents across the cut.

The functional consequences of this morphological continuity and elemental retention are quantified electrically. The conductivity of the healed PGNCM gel provides a functional measure of conductive-network recovery across the cut. The pristine PGNCM hydrogel exhibits a conductivity of 0.296 S/m. During the first F–T healing cycle, the conductivity progressively increased to 0.128 ± 0.006 , 0.174 ± 0.026 , and 0.247 ± 0.015 S/m after healing durations of 12, 18, and 24 h, respectively, corresponding to approximately 43, 59, and 83 % of the pristine value (Fig. 4c). After a second 24 h F–T cycle, the conductivity further increased to 0.258 ± 0.025 S/m, corresponding to 87 % recovery. These results demonstrate a clear healing-duration-dependent restoration of the conductive pathway. The healing-duration dependence was also evaluated mechanically using tensile tests after two

successive F–T healing cycles (Fig. S11). As the freezing duration per cycle increased from 12 to 24 h, the tensile strength increased from 17.19 ± 0.81 to 33.22 ± 5.69 kPa, the elongation at break from 62.6 ± 3.9 to 109.6 ± 7.0 %, and the toughness from 5.45 ± 0.53 to 15.93 ± 3.24 kJ/m³, while Young's modulus remained within a comparable range of 24.2–29.7 kPa. These results demonstrate progressive mechanical reintegration of the cut interface with increasing F–T healing duration.

To evaluate functional durability after healing, the pristine and self-healed PGNCM sensors were compared under the same long-term cycling protocol of 1000 loading–unloading cycles at 25 % strain (Fig. 4d). The pristine sensor maintained a regular cyclic response throughout the test, whereas the self-healed sensor retained repeated cyclic modulation despite a gradual baseline shift. The representative early- and late-stage traces further show that the cyclic response remained clearly resolved after healing. These results indicate that the self-healed sensor retained substantial electromechanical functionality under repeated 25 % deformation. Across the mechanical, electromechanical sensing, and healing metrics characterized in Sections 2.2–2.4, Table S2 benchmarks PGNCM against representative PVA-based

hydrogels and highlights its balanced combination of tensile strength, stretchability, strain sensitivity, conductivity recovery, and post-healing cycling durability.

2.5. Sensing performance in diverse aqueous environments

The sensing performance in Section 2.3 was obtained from the as-prepared gel in air. Subsequently, we examined whether this response was preserved after prolonged immersion in different aqueous media and during operation at different water-bath temperatures. The dimensional-stability data in Section 2.1 showed that PGNCM exhibited a swelling ratio of 53.99 % after 48 h in artificial seawater, which was the lowest among the tested formulations. This reduced swelling was consistent with previous reports on swelling-resistant hydrogel strain sensors designed for underwater operation. Throughout this work, swelling resistance (the reduced mass uptake measured over 48 h in artificial seawater), short-term underwater functional stability (the retention of mechanical integrity and sensing response during 2 days of

immersion and underwater operation), and long-term seawater durability (dimensional and electromechanical stability beyond the tested immersion window) are treated as distinct properties. The measurements reported below address the first two.

The electrical continuity under direct submersion was evaluated (Fig. 5a, Movie S2). The PGNCM gel was wired as the sole conductive bridge of an external circuit driving an LED array. The resulting illumination provided a directly visible optical readout of bridge conductivity. The illumination pattern was preserved without a perceptible loss of brightness when the assembly was transferred from air into a DIW bath, indicating that the conductive network identified in Section 2.3 (Fig. 3a) remained electrically continuous under direct aqueous contact.

The responses to different bending angles and frequencies were evaluated using a finger-mounted sensor (Fig. 5b, Movie S2). Under a stepwise increase in the bending angle from 0° to 90° at a fixed bending rate (Fig. 5c, top), the sensor produced well-defined plateaus whose peak amplitudes increased with bending angle, consistent with the strain-dependent resistance response established in Section 2.3 (Fig. 3b

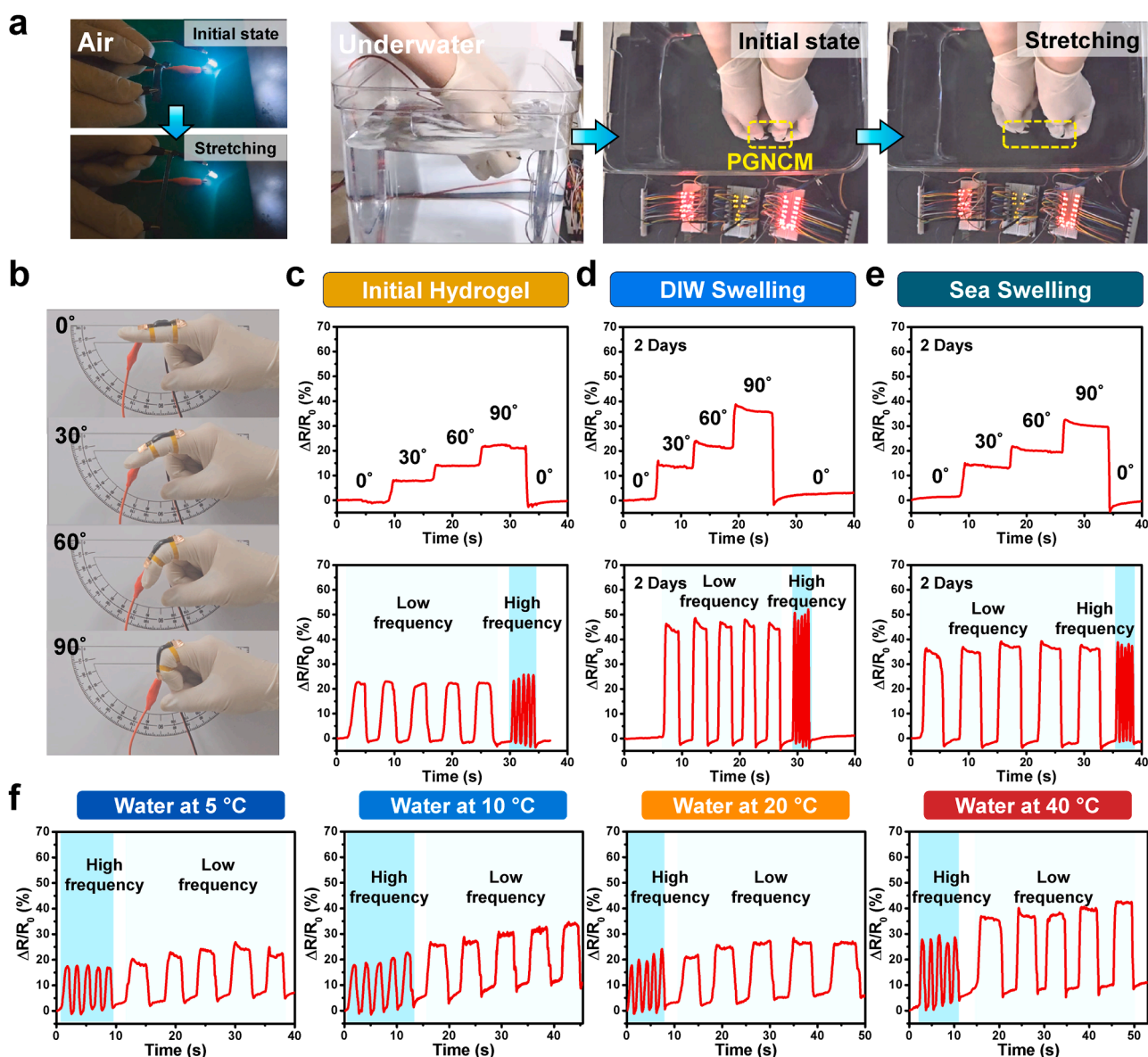


Fig. 5. Underwater sensing performance of the PGNCM sensor in diverse aqueous environments. (a) LED-circuit demonstration of electrical continuity in air and underwater. (b) Digital photographs of the finger-mounted sensor at bending angles from 0° to 90°. $\Delta R/R_0$ responses versus bending angle and bending frequency for (c) the as-prepared sensor, (d) the sample after two days of DIW immersion, and (e) the sample after two days of artificial-seawater immersion. (f) $\Delta R/R_0$ traces of the underwater sensor at water temperatures of 5, 10, 20, and 40 °C.

and e). When the bending angle was fixed, the sensor was manually bent at low and high frequencies (Fig. 5c, bottom). The peak-to-peak amplitude of $\Delta R/R_0$ remained comparable under both conditions, while the cycle period changed with the bending frequency. This behavior complemented the air-side stretching rate-dependent measurements (Fig. S10).

The same bending angle and frequency protocol was repeated on samples immersed for two days in DIW (Fig. 5d) and artificial seawater (Fig. 5e). Before measurement, the sensors were removed from the respective immersion media. Comparable responses were observed after immersion in both media. This preservation was consistent with the reduced 48 h swelling observed in Section 2.1 and the bulk electrical conductivity characterized in Section 2.3. Therefore, the sensing

behavior was largely retained despite immersion in media with different ionic compositions, which was consistent with the results of previous reports on anti-swelling hydrogel sensors [56,67].

The remaining environment condition, i.e., water-bath temperature, was varied at 5, 10, 20, and 40 °C (Fig. 5f), spanning a range relevant to laboratory and near-surface aquatic environments. At the four bath temperatures, $\Delta R/R_0$ traces exhibited comparable peak-to-peak amplitudes and periodic signal profiles. This thermal robustness suggested that the freeze-thaw-derived PVA framework and dynamic physical interactions remained sufficiently stable throughout the tested range (Sections 2.1 and 2.2).

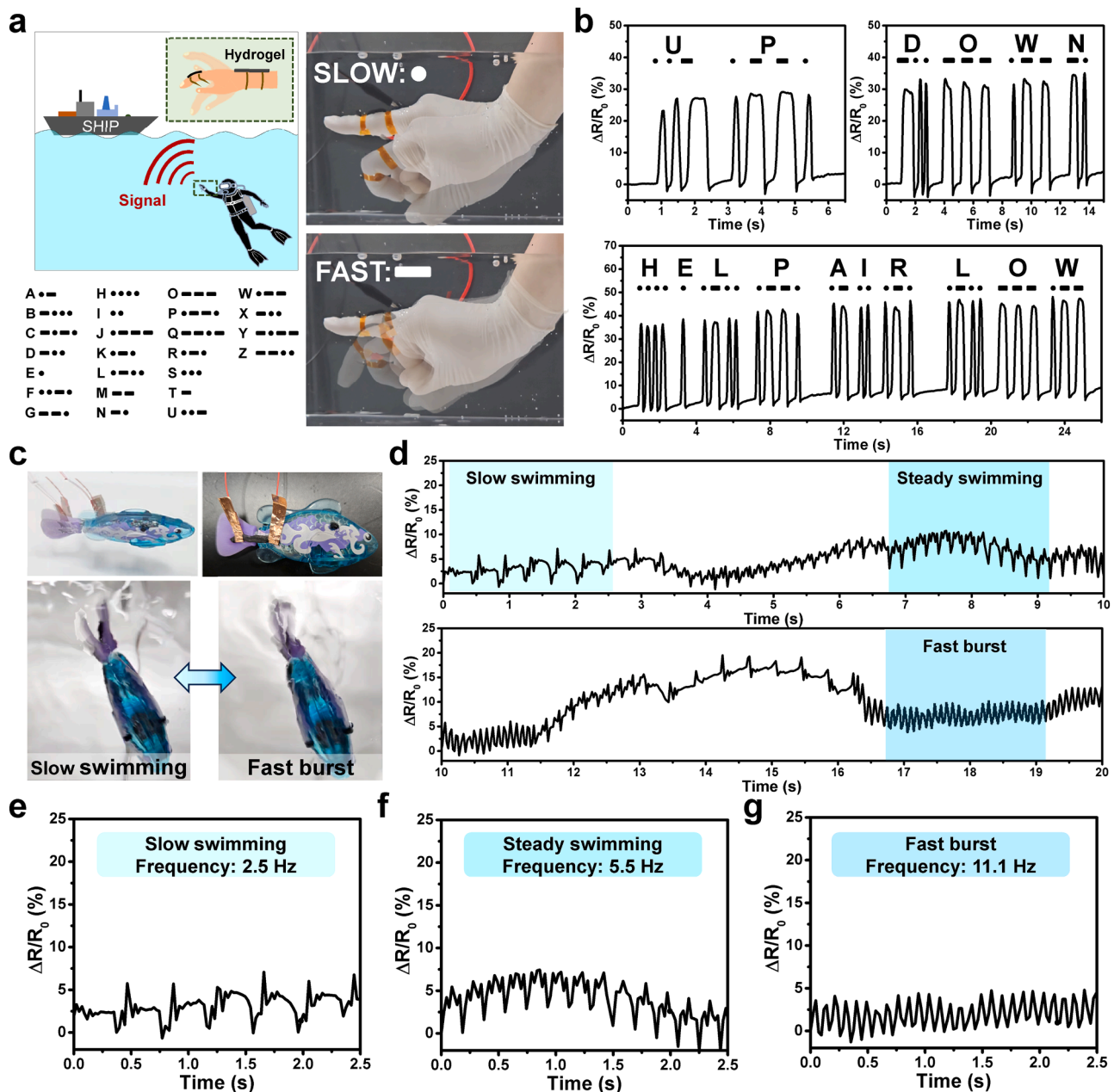


Fig. 6. Underwater communication and robotic-fish sensing demonstrations. (a) Schematic of the underwater communication scheme with the Morse code reference chart. (b) Real-time decoding of underwater finger-bending inputs into the standard Morse-encoded words. (c) Optical images of the PGNCM-integrated motor-driven robotic fish in two locomotion modes: slow swimming (left) and fast burst (right); the burst mode beats the tail at a markedly shorter stroke period, which is then transduced into the higher-frequency $\Delta R/R_0$ response resolved in Fig. 6e and g. (d) Continuous $\Delta R/R_0$ trace recorded across an extended swimming sequence covering several locomotion modes. Magnified time-resolved signals with the tail-beat frequency obtained by peak counting indicated on each panel: (e) slow swimming at 2.5 Hz, (f) steady swimming at 5.5 Hz, and (g) fast burst at 11.1 Hz.

2.6. Demonstration of underwater communication and robotic-fish sensing

Based on the sensing performance established in Sections 2.3–2.5, including linearity ($GF = 1.27$, $R^2 = 0.997$), cyclic stability, and underwater sensing performance, two functional demonstrations were conducted on a single-sensor element without covalent modification or hydrophobic encapsulation. The sensor maintained sub-half-second response and recovery times and retained its strain response after two days of immersion in DIW and artificial seawater, as well as at water-bath temperatures in the range of 5–40 °C. The demonstrations consisted of real-time underwater Morse code communication and the tracking of a motor-driven robotic fish.

Underwater Morse code transmission was implemented by mounting the PGNCM hydrogel sensor on the dorsal side of the index finger and submerging the assembly in an artificial-seawater bath (Fig. 6a). Short and long finger bends were mapped to dots and dashes according to the international Morse code reference chart shown alongside the schematic. The operator transmitted five short status words, and the $\Delta R/R_0$ trace was decoded in real time at the receiver (Fig. 6b). Each dot and dash produced a well-defined peak that returned to a stable baseline between successive elements, with no observable drift during the 26 s transmission. At the natural cadence of the operator, the dot and dash signals remained readily distinguishable in time. This is consistent with the 306 ms response time and 345 ms recovery time established in Section 2.3 (Fig. 3f), both of which are well below the spacing between successive Morse elements. A full five-word sequence was transmitted, received, and decoded without character ambiguity, demonstrating the feasibility of underwater signal transmission using the PGNCM sensor. Morse-code-based information transmission using mechanically induced electrical signals has also been demonstrated in various multifunctional gel-based sensors [15–17,59–61]. Morse-code-based underwater communication has been previously demonstrated in several hydrogel systems [50,57,67–69].

The second demonstration extends the sensor to monitoring robotic-fish locomotion. To ensure that each tail beat imposed a defined bending strain on the gel, the PGNCM hydrogel strip was externally bonded across the body-tail junction onto the dorsal surface at the caudal-peduncle region of a tail-actuated, motor-driven robotic fish (Fig. 6c, Movie S3). Air and underwater controls at matched drive frequencies (Movie S4) produced comparable peak shapes and periods, suggesting that the signal originated predominantly from tail-beat-imposed bending. Under the same submerged drive conditions, the $\Delta R/R_0$ signal was recorded continuously while the actuator stepped sequentially through slow-swimming, steady-swimming, and fast-burst modes (Fig. 6d). Corresponding real-time recordings are provided in Movie S4, which starts with the in-air control and progresses through three submerged modes. The continuous trace exhibited an oscillatory $\Delta R/R_0$ response throughout the sequence, with three distinct regimes that differed in peak rate and waveform while maintaining a stable baseline.

Representative segments corresponding to the three locomotion modes were analyzed separately (Fig. 6e–g). The $\Delta R/R_0$ signal remained regular and periodic in the slow-, steady-, and fast-burst modes (Fig. 6e–g). The peak-counted tail-beat frequency increased from 2.5 Hz in the slow-swimming mode to 5.5 and 11.1 Hz in the steady-swimming and fast-burst modes, respectively, with a decrease in the oscillation period. Across the three modes, the measured tail-beat frequency tracked the actuator-imposed frequency, corresponding to a 4.4-fold increase. Multimode robotic-fish tracking has previously been reported using hydrogel strain sensors [69,70]. These sensors quantitatively tracked tail-beat frequency under direct underwater exposure across all three swimming modes.

3. Conclusion

This study established the PGNCM hydrogel as a fully physical, dual-

salt PVA hydrogel that integrates reduced saline swelling, conductive self-healing, and underwater strain sensing within a single-material platform. The complementary roles of NaCl and CaCl_2 were proposed to facilitate salting-out-assisted network ordering and divalent ion-mediated interactions within the PVA/fMWCNT network, thereby enabling the formation of a mechanically robust and conductive network without covalent crosslinkers, zwitterionic comonomers, or hydrophobic encapsulation. Consequently, the hydrogel exhibited a swelling ratio of ~54 % after 48 h in artificial seawater and recovered 87 % of its pristine conductivity after freeze-thaw-assisted healing. However, long-term seawater immersion and healing under ambient conditions are yet to be addressed.

The PGNCM hydrogel strain sensor provided a linear electromechanical response over 0–200 % strain ($GF = 1.27$, $R^2 = 0.997$), allowed sub-half-second response and recovery, and retained repeated resistance modulation over 1000 loading cycles at 100 % strain, with ~89 % retention of the peak-to-peak response amplitude despite baseline drift. This sensing behavior was largely retained after two days of immersion in DIW and artificial seawater and over a water-temperature range of 5–40 °C. These characteristics enabled underwater Morse code transmission and robotic-fish locomotion tracking upon direct aqueous exposure using the same sensor element. These results demonstrate that a fully physical, dual-salt-assisted freeze-thaw strategy provides a practical design route toward hydrogel electronics for underwater sensing, communication, and soft robotic systems.

4. Experimental section

4.1. Materials

PVA ($M_w = 126,000$, 98 % hydrolyzed) was purchased from Scientific Polymer Products, Inc., USA. Glycerol (glycerin, purity ~ 99 %) and calcium chloride (CaCl_2 , purity ~ 93 %) were purchased from Daejung Chemicals & Metals Co., Ltd., Korea. Sodium chloride (NaCl, purity ~ 99 %) was obtained from Duksan Pure Chemicals Co. Ltd., Korea. Carboxyl-fMWCNTs (10–20 nm outside diameter, 5–10 nm inside diameter, 10–30 μm length, ~95 % purity, >100 S/cm electrical conductivity) were purchased from US Research Nanomaterials Inc. and used as the conductive filler. Pristine MWCNTs (pMWCNTs, 10–20 nm outside diameter, 5–10 nm inside diameter, 10–30 μm length, ~95 % purity, >100 S/cm electrical conductivity) were purchased from US Research Nanomaterials Inc. and used as the non-functionalized MWCNT control. Artificial seawater was prepared for swelling and underwater sensing tests by dissolving commercial sea salt (TARA SPS Coral Sea Salt, Ace Korea, China) in DIW. The chemical composition of sea salt includes major ions such as Na^+ (9600–9900 mg/L), Cl^- (18,000–19,300 mg/L), Mg^{2+} (1400–1550 mg/L), and Ca^{2+} (440–480 mg/L), mimicking the environment of natural seawater.

4.2. Preparation of PGNCM hydrogels

The PGNCM hydrogels were synthesized via a one-pot heating-stirring method followed by a cyclic freeze-thaw process. First, 1 g of NaCl and 1 g of CaCl_2 were dissolved in 40 g of DIW to form a saline precursor solution. Subsequently, 4 g of PVA (10 wt% relative to DIW) and 2 g of glycerol were added to the solution. The mixture was stirred at 600 rpm and heated on a hotplate set to 180 °C until the PVA powder was completely dissolved, resulting in a clear and viscous solution. fMWCNTs were first dispersed in DIW at 1 wt % using an ultrasonicator (Q700 Sonicator, Qsonica Co.) at an intensity of 60 W for 20 min to obtain a uniform fMWCNT dispersion. Then, 1 mL of the fMWCNT aqueous dispersion was added to the PVA/glycerol/salt mixture and stirred for an additional 1 h to ensure uniform dispersion. Physical crosslinking was induced through two successive freeze-thaw cycles, where the samples were frozen at –18 °C for 24 h and thawed at room temperature for 1 h. For comparison, PG, PGN, PGC, and PGNC

hydrogels were prepared following the same procedure and selectively omitting NaCl, CaCl₂, and fMWCNTs. For the single-salt controls, PGN and PGC contained 2 g of NaCl and CaCl₂, respectively, to maintain the same total salt content as PGNCM. Additional control formulations were prepared using the same procedure. PGM consisted of PVA/glycerol/fMWCNTs without salts, PGNM contained PVA/glycerol/NaCl/fMWCNTs without CaCl₂, and PGNC-pM was prepared using non-carboxylated MWCNTs in place of fMWCNTs in the dual-salt formulation. PNCM was prepared by omitting glycerol from the PGNCM formulation. The detailed compositions of all hydrogel formulations are summarized in Table S4.

4.3. Characterization

SEM (Quattro ESEM, Thermo Scientific) was employed to characterize the morphology of the PGNCM hydrogel. The molecular structures and chemical groups present in the hydrogels were analyzed using a Fourier-transform infrared spectrometer (Nicolet iS50). The electrical responses of the hydrogel sensor were monitored using a digital source meter (SMU 2611B, Keithley Instruments, Ltd.) by applying a voltage of 0.1 V at ~25 °C under ambient conditions. X-ray diffraction (XRD) measurements were performed on freeze-dried hydrogel samples using a MiniFlex 600 diffractometer (Rigaku) operated at 40 kV and 15 mA. The diffraction patterns were collected over a 2θ range of 5–60° with a step size of 0.02° using continuous scanning. X-ray photoelectron spectroscopy (XPS) was performed on freeze-dried hydrogel samples using a Thermo Scientific K-Alpha+ spectrometer with monochromated Al Kα radiation (1486.6 eV) and a 300 μm analysis spot. Survey and high-resolution spectra were acquired at pass energies of 200 and 50 eV with step sizes of 1.0 and 0.1 eV, respectively. Charge compensation was applied during acquisition. The spectra were analyzed using CasaXPS and charge-referenced to the low-binding-energy C 1 s component, predominantly assigned to C–C/C–H, at 284.8 eV. Shirley backgrounds and LA(50) line shapes were employed for peak fitting. Differential scanning calorimetry (DSC, DSC 214, NETZSCH) was performed by cooling the samples from 25 to –60 °C and subsequently heating them from –60 to 40 °C at a scanning rate of 10 °C/min.

4.4. Swelling and moisture-retention behavior

The swelling behavior of the hydrogels was evaluated through immersion tests conducted in DIW and artificial seawater. The artificial seawater was prepared by dissolving commercial sea salt in purified water at a mass-to-volume ratio of 33.3 g/L. The resulting solution was aerated to achieve oxygen/carbon dioxide equilibrium, yielding a salinity of 33.0–35.0 ppt, specific gravity of 1.023–1.025, and pH of 8.30–8.50 at 25 °C. Initial as-prepared hydrogel samples were immersed in the corresponding media at room temperature. At specific time intervals, the samples were removed, lightly dabbed with a filter paper to eliminate excess surface moisture, and weighed. The swelling ratios were calculated using

$$\text{Swelling ratio} = \frac{W_s - W_0}{W_0} \times 100 \%,$$

where W_0 and W_s represent the weights of the hydrogel before and after swelling, respectively.

For the moisture-retention test, PNCM and PGNCM hydrogels ($n = 3$) were weighed at predetermined intervals during storage for 7 days. The weight retention was calculated as $W_t/W_0 \times 100$, where W_0 and W_t represent the initial weight and the weight at time t , respectively.

4.5. Mechanical performance

The mechanical properties of the hydrogels were investigated using an electronic universal testing machine (MCT-2150, A&D Co.) equipped

with a 500 N load cell at room temperature. For standardized characterization, the tensile samples were fabricated by infusing the precursor solution into three-dimensional-printed molds that complied with ASTM D638 specifications. Uniaxial tensile tests were performed at a constant stretching rate of 100 mm/min. For the stretching rate-dependent measurements, additional tensile tests were performed at stretching rates of 50, 100, and 200 mm/min. Force and displacement data were synchronized to establish comprehensive stress–strain relationships. Young's modulus was derived from the slope of the stress–strain curve within the initial linear region (5–15 % strain), whereas the toughness was determined by integrating the area under the stress–strain curve. Cyclic compression tests were performed on cylindrical specimens for 20 loading–unloading cycles to 50 % strain at a compression rate of 100 mm/min at room temperature.

4.6. Viscoelastic performance

The viscoelastic properties of the hydrogels were characterized using a rotational rheometer (MCR 702e, Anton Paar, Austria) equipped with a 25-mm-diameter parallel-plate (PP25) geometry. All measurements were conducted at a fixed gap of 1.0 mm and under a controlled temperature of 25 °C. The linear viscoelastic region was determined by performing amplitude sweep tests at a fixed frequency of 10 rad/s with the shear strain varying over 0.1–100 %. Subsequently, frequency sweep tests were conducted within the identified linear viscoelastic region at a constant strain of 0.1 % over a frequency range of 0.1–100 rad/s.

4.7. Conductivity measurement

The electrical properties of the PGNCM hydrogel were evaluated using a digital source meter (SMU 2611B, Keithley Instruments, Ltd.). The electrical conductivity (σ , S/m) was calculated using

$$\sigma = \frac{L}{RS},$$

where R represents the resistance value (Ω), and L and S represent the length between electrodes (m) and sectional area of the hydrogel (m²), respectively. For the healing-duration-dependent measurements, completely cut PGNCM specimens were brought into contact and frozen at –18 °C for 12, 18, or 24 h, followed by thawing at room temperature for 1 h before conductivity measurement. For the two-cycle condition, the specimens were subjected to a second 24 h freezing step followed by the same thawing procedure.

4.8. Strain-sensing performance

The strain-sensing capabilities of the hydrogel were evaluated by fabricating it into a strain sensor by affixing copper wires to both endpoints, followed by wrapping with copper foil to ensure stable electrical contact. This sensor, which had a uniform thickness of ~2 mm, was then subjected to mechanical deformation. The real-time resistance (R_t) was monitored through amperometric R – t curves at a constant voltage. The relative change in resistance was determined using

$$\frac{\Delta R}{R_0} = \frac{R_t - R_0}{R_0},$$

where R_0 represents the original resistance without applied strain. The sensitivity of the sensor was quantified using the GF defined as

$$GF = \frac{\Delta R/R_0}{\epsilon}, \text{ where } \epsilon \text{ represents the applied longitudinal strain.}$$

Electrical hysteresis was evaluated from a single loading–unloading cycle to 100 % strain and calculated as the area enclosed between the loading and unloading $\Delta R/R_0$ curves normalized by the area under the loading curve. For the low-strain measurements, cyclic tensile strains of 1–5 % were applied. Long-term cyclic durability was evaluated over 1000 loading–unloading cycles at 100 % strain. Device-to-device

reproducibility was assessed using three independently fabricated PGNCM sensors, and the GF values are reported as the mean $\pm$ standard deviation ($n = 3$).

For application-level sensing tests, the PGNCM sensor was mounted on the dorsal side of the index finger using Kapton tape and evaluated at different bending angles and manually varied low and high frequencies. After 2 days of immersion in DIW or artificial seawater, the sensors were removed from the media and evaluated using the same bending protocol. Underwater Morse-code measurements were performed while the finger-mounted sensor remained submerged in artificial seawater. For robotic-fish sensing, the sensor was attached across the dorsal body–tail junction using an adhesive, and tail-beat frequencies were determined by peak counting of the resistance signals.

CRediT authorship contribution statement

Seonghoon Lee: Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Keuntae Baek:** Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Animesh Sinha:** Methodology, Investigation, Conceptualization. **Hyeyun Kang:** Methodology, Investigation. **Hongyun So:** Writing – review & editing, Supervision, Funding acquisition, Conceptualization.

Declaration of competing interest

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

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at [doi:10.1016/j.snr.2026.100504](https://doi.org/10.1016/j.snr.2026.100504).

Data availability

Data will be made available on request.

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