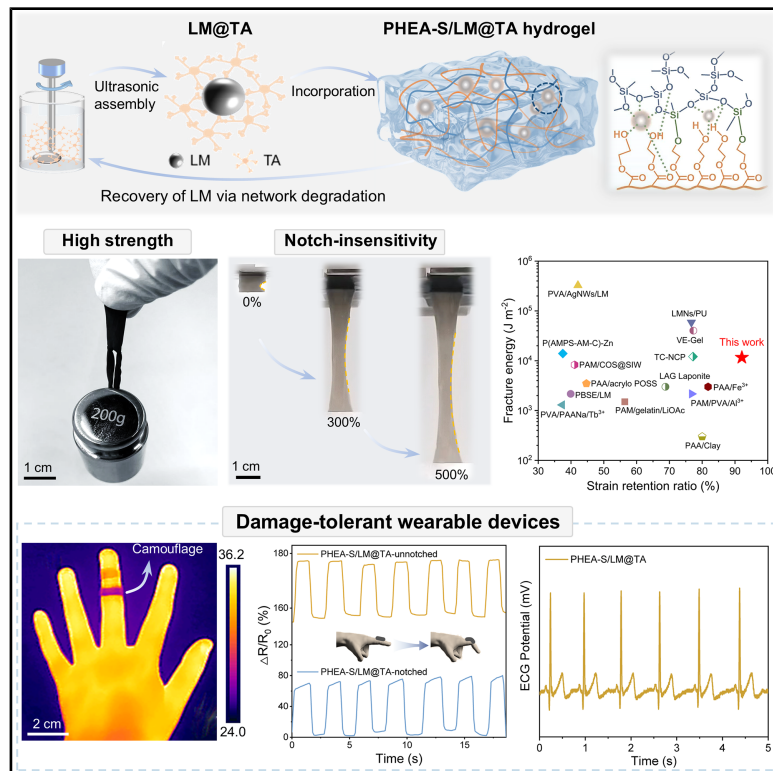


Interface-engineered liquid metal-composited hydrogels with high toughness and extreme flaw insensitivity for damage-tolerant wearables

Graphical abstract



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In brief

This work proposes an interface-engineered strategy by coating liquid metal (LM) with tannic acid to stabilize LM dispersion in aqueous media and establish strong yet dynamic interactions with the hydrogel matrix. The resulting LM-composited hydrogels achieve high stretchability, toughness, and exceptional flaw insensitivity, providing a versatile platform for damage-tolerant wearable devices.

Highlights

- Tannic acid coating transforms liquid metal droplets into active crack-blunting fillers
- Dynamic filler-matrix coupling enables synchronized deformation and crack-tip blunting
- The hydrogel achieves high toughness and extreme flaw insensitivity
- Damage-tolerant wearable devices enable reliable flexible sensing and IR camouflage



Development

Practical, real world, technological considerations and constraints

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Article

Interface-engineered liquid metal-composited hydrogels with high toughness and extreme flaw insensitivity for damage-tolerant wearables

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PROGRESS AND POTENTIAL Functional filler-composited hydrogels have emerged as promising materials for flexible and wearable devices, yet their practical reliability remains constrained by flaw sensitivity. Even small defects can trigger crack propagation, mechanical failure, and loss of functionality, limiting long-term reliability. While soft liquid metal (LM) nanodroplets have been considered as deformable fillers capable of alleviating stress concentration, their high surface tension and weak interactions with polymer networks have prevented the realization of highly flaw-insensitive LM-composited hydrogels. In this work, we report that polyphenol-mediated interfacial engineering can transform LM nanodroplets from passive fillers into active crack-resistant components. Tannic acid coating on LM nanodroplets not only stabilize LM dispersions in aqueous media but also establish strong yet dynamic interactions with the hydrogel matrix, enabling efficient stress transfer and crack-tip blunting via synchronized deformation between the soft LM fillers and the polymer network. This resulting LM-composited hydrogels combine high stretchability, toughness, and exceptional flaw insensitivity, which supports reliable sensing and photothermal functionalities even after part mechanical damage. More broadly, this work highlights the critical role of polyphenol-mediated interfacial design in governing the mechanical behavior of soft composites. The concept of transforming soft fillers into active stress-management units may open new opportunities for designing damage-tolerant materials for wearable electronics, soft machines, and other emerging technologies.

SUMMARY

Achieving highly flaw-insensitive liquid metal (LM)-composited hydrogels for damage-tolerant wearable devices remains challenging due to the high surface tension of LM and weak filler-matrix interactions. Here, we demonstrate that tannic acid (TA) coating on LM nanodroplets (LM@TA) addresses these challenges by stabilizing LM dispersion and establishing strong yet dynamic interactions with a semi-interpenetrating poly(hydroxyethyl acrylate)-siloxane (PHEA-S) network. This interfacial engineering enables synchronized deformation between LM@TA fillers and the polymer matrix, effectively blunting crack tips and delocalizing stress. The resulting hydrogels exhibit high stretchability (860%), tensile strength (0.75 MPa), and toughness (3.09 MJ m^{-3}), while achieving exceptional flaw-insensitivity (fracture strain retention of up to 92% and fracture energy of 12.43 kJ m^{-2}). Notably, the hydrogel enables crack-tolerant wearable sensors for reliable motion and electrophysiological monitoring, while the plasmonic LM nanodroplets impart photothermal functionality for infrared camouflage, highlighting the broad potential of this polyphenol-mediated interfacial engineering strategy for damage-tolerant flexible electronics.

INTRODUCTION

Hydrogels, featuring skin-like softness, high ductility, and good biocompatibility, have been widely explored as flexible wearable

devices for health monitoring, human-machine interaction, and soft robotics.^{1–5} To meet functional demands such as conductivity, sensing, and photothermal response, a wide variety of fillers have been incorporated into hydrogel networks.^{6–9} For

long-term wearable use, however, the relevant performance target is not how strong a pristine hydrogel can be, but how reliably it functions after defects appear. In practice, repeated deformation inevitably introduces such defects, ranging from surface scratches and punctures to fatigue-induced micro-cracks.^{10–12} However, conventional rigid fillers exacerbate this vulnerability by introducing mechanical mismatch at filler-matrix interfaces, leading to stress concentration at crack tips and accelerating crack propagation from pre-existing defects.¹³ While considerable progress has been made in enhancing bulk and fracture toughness through double-network architectures and sacrificial bond designs, high fracture toughness does not necessarily translate into high flaw insensitivity.^{14–16} When cracks are present, these hydrogels typically exhibit largely reduced fracture strain relative to their pristine state and undergo premature failure.¹⁷ This limitation is particularly distinct for filler-composited hydrogels, where failure is governed by defect-induced crack propagation at the filler-matrix interface.^{18,19} Consequently, developing filler-composited hydrogels that are not only tough but also highly flaw-insensitive remains a pressing challenge for achieving reliable long-term operation.

Recent works have shown that liquid metal (LM) nanodroplets offer a fundamentally different reinforcement paradigm.^{20–24} Unlike rigid nanofillers that concentrate stress at filler-matrix interfaces, soft LM droplets can deform in concert with the surrounding polymer network, thereby avoiding the mechanical mismatch that limits conventional composites and effectively blunting crack propagation.^{25–29} Indeed, LM-composited hydrogels have already achieved impressive stretchability exceeding 1,400% and conductivity above $1,800 \text{ S cm}^{-1}$.³⁰ However, these efforts have focused almost exclusively on bulk mechanical enhancement and electrical performance. The development of highly crack-resistant LM-composited hydrogels has been rarely studied. This gap originates from the high surface tension of LM droplets in aqueous environments and the weak interfacial interactions between LM droplets and polymer network.^{31,32} From a fracture mechanics perspective, two key conditions must be satisfied to achieve effective crack resistance. First, a stable and homogeneous dispersion of fillers within the polymer matrix is essential for uniform stress distribution and dissipation.^{28,33} Second, strong yet dynamic interfacial coupling between the fillers and the polymer network is required to enable coordinated deformation and effective energy dissipation at the crack front.³⁴ Although tannic acid (TA) coatings have been widely used to stabilize LM dispersions for the fabrication of adhesive LM inks,³⁵ conductive hydrogel adhesives,²⁰ lubricating additives,³⁶ conductive yarns,³⁷ and adhesive hydrogel electrodes,³⁸ and crack resistance in LM-composited hydrogels has been pursued through structural design,³⁹ the mechanical role of polyphenol-mediated interfaces in governing fracture behavior remains largely unexplored. In most existing hydrogels, LM nanodroplets are merely trapped within the network through weak physical interactions, limiting their ability to transfer stress or participate in energy dissipation near crack tips. These limitations highlight a clear design principle for highly flaw-insensitive LM-composited hydrogels: the filler-matrix interface must be sufficiently strong to transfer stress under elastic deformation, yet sufficiently dynamic to rupture sequentially at the crack tip and dissipate

energy before catastrophic propagation. Polyphenolic hydrogen bonds, with bond energies on the order of 5–30 kJ/mol,⁴⁰ sit precisely in this regime, much weaker than covalent crosslinks but much stronger than van der Waals contacts, making them well suited to mediate this dual function.

Guided by this principle, here we demonstrate that polyphenol-mediated interfacial engineering can transform LM nanodroplets from passive inclusions into active participants in crack resistance. In our system, TA-stabilized LM nanodroplets (LM@TA) are incorporated into a semi-interpenetrating poly(hydroxyethyl acrylate)-siloxane (PHEA-S) network. TA acts as a dual-functional interfacial mediator: its catechol groups coordinate with the gallium oxide surface of LM, forming a stable coating that suppresses aggregation, while its abundant phenolic hydroxyl groups establish extensive hydrogen-bonding interactions with the PHEA-S network, thereby creating a robust yet dynamic filler-matrix interface. This engineered interface enables synchronized filler-matrix deformation that effectively blunts crack tips and dissipates energy across multiple length scales. The resulting hydrogel achieves an outstanding fracture energy of 12.43 kJ m^{-2} and retains up to 92% of its fracture strain, depending on notch geometry, at a notch-to-width ratio of 0.4, while also exhibiting high stretchability (860%), tensile strength (0.75 MPa), and toughness (3.09 MJ m^{-3}). Leveraging these advantages, we demonstrate that wearable devices fabricated from this hydrogel deliver stable sensing performance and achieve dynamic infrared camouflage even after crack formation, enabling reliable monitoring of human motion under damage conditions and electrophysiological signals. Furthermore, the LM embedded in spent hydrogels can be efficiently recovered by degrading the crosslinked polymer network in NaOH solution. This work establishes polyphenol-mediated interfacial engineering as a design principle for damage-tolerant soft-filler hydrogels, with potential extension to broader soft-filler chemistries.

RESULTS

TA-mediated stabilization of LM nanodroplets and their integration into semi-IPN hydrogels

LM@TA nanodroplets were prepared by sonicating bulk eutectic gallium-indium (EGaIn) in an ethanolic TA solution, during which TA molecules rapidly anchored onto freshly generated LM nanodroplet surfaces (Figure 1A). High-resolution transmission electron microscopy (HRTEM) revealed that the resulting LM@TA nanodroplets exhibited a uniform spherical morphology with an average diameter of approximately 400 nm (Figure S1). Energy-dispersive X-ray spectroscopy (EDS) mapping confirmed the homogeneous spatial distribution of C, O, Ga, and In elements across individual nanodroplets, suggesting a conformal TA coating on the LM surface rather than physical adsorption.

To elucidate the chemical nature of the TA-LM interface, we employed Fourier transform infrared (FTIR) spectroscopy and X-ray photoelectron spectroscopy (XPS). In the FTIR spectra (Figure S2), characteristic peaks of TA, including phenolic C–O stretching ($1,531$ and $1,441 \text{ cm}^{-1}$) and O–H bending ($1,308$ and $1,180 \text{ cm}^{-1}$), shifted to lower wavenumbers ($1,488$, $1,358$, $1,231$, and $1,154 \text{ cm}^{-1}$) in LM@TA, indicative of electron density

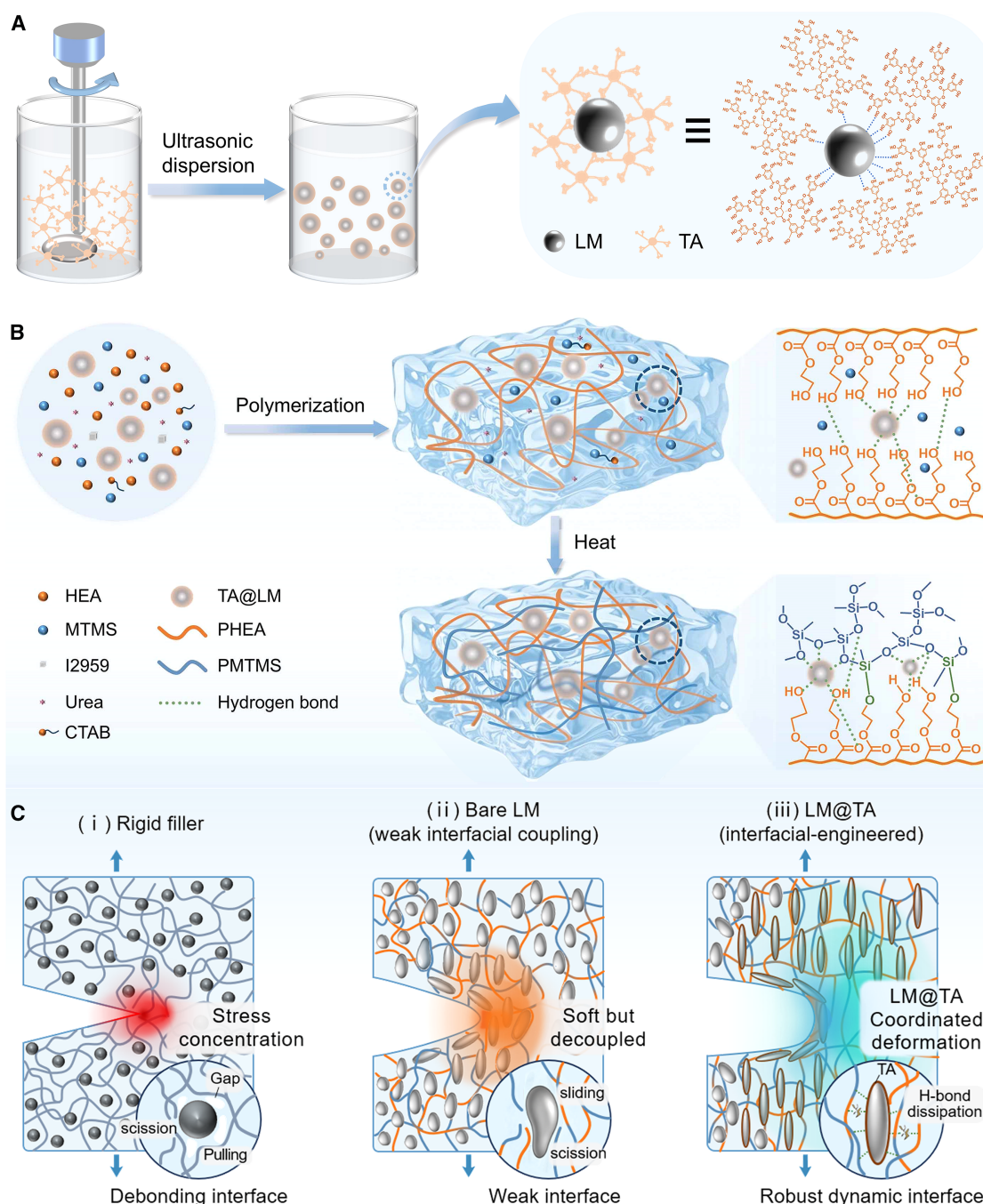


Figure 1. Fabrication and structural design of LM@TA nanodroplets and the PHEA-S/LM@TA hydrogel

(A) Schematic illustration of LM@TA preparation via ultrasonic dispersion with TA surface modification.

(B) Schematic of the PHEA-S/LM@TA hydrogel fabrication, highlighting the semi-IPN structure and multiple hydrogen-bonding interactions between LM@TA and polymer networks.

(C) Schematic illustration of crack-tip responses in hydrogels with distinct filler-matrix coupling states.

redistribution upon coordination between catechol groups and metal species.^{35,38} XPS analysis provided direct evidence for this metal-catechol coordination (Figure S3). In the Ga 3d spectrum, the binding energies of both Ga^{3+} and Ga^0 shifted to higher values upon TA modification (from 19.53 to 20.16 eV and from

17.60 to 18.00 eV, respectively), confirming the formation of Ga–O–C coordination bonds.⁴¹ A similar positive shift was observed in the In 3d spectrum, accompanied by the transition of In^0 to In^{3+} , indicating that both gallium and indium oxide species on the LM surface participate in coordination with TA.³⁵

Additionally, the emergence of a new peak at 531.96 eV in the O 1s spectrum, attributed to C–O–Ga bonding, further corroborates the covalent anchoring of TA onto the LM surface.⁴¹ These spectroscopic results collectively demonstrate that TA is robustly anchored onto the LM nanodroplet surface through metal-catechol coordination, establishing a chemically stable interface that serves as the foundation for subsequent dispersion stabilization and filler-matrix bonding.

The robust TA coating fundamentally altered the surface properties of LM nanodroplets, enabling their stable dispersion in aqueous environments. Static water contact angle measurements revealed a dramatic decrease from 55.9° for pristine LM to 14.9° for LM@TA (Figure S4), indicating that the TA coating transformed the LM surface from a moderately hydrophobic to superhydrophilic state. This enhanced hydrophilicity arises from the abundant phenolic hydroxyl groups on TA, which significantly reduce the interfacial tension between LM nanodroplets and the surrounding aqueous medium. Dynamic light scattering (DLS) analysis further demonstrated the stabilizing effect of TA modification (Figure S5). While bare LM nanodroplets exhibited a broad size distribution centered at approximately 615 nm, LM@TA nanodroplets displayed a markedly narrower distribution with a reduced average diameter of 395–400 nm at LM-to-TA mass ratios of 2:1–10:1, consistent with the TEM observations. However, increasing the LM-to-TA mass ratio to 20:1 and 30:1 led to enlarged LM@TA droplet sizes (~460 nm), indicating that moderate TA coverage is essential for stabilizing LM@TA nanodroplets, whereas excessively low TA content is insufficient to suppress droplet coalescence, resulting in particle growth. The long-term colloidal stability was assessed through sedimentation tests (Figure S6). Pristine LM droplets rapidly aggregated and settled within 2 h, producing a clarified supernatant with substantial metal deposits at the bottom due to their intrinsically high surface tension.³⁸ For LM@TA dispersions prepared with low TA contents (LM-to-TA mass ratios of 20:1 and 30:1), part sedimentation was observed after storage at room temperature for 24 h. In contrast, LM@TA nanodroplets prepared at an LM-to-TA mass ratio of 10:1 formed a homogeneous gray-black colloidal dispersion that remained stable without visible sedimentation even after 24 h. These results collectively confirm that moderate TA coating fulfills the first key function of our interfacial design: stabilizing LM nanodroplets against aggregation, thereby ensuring their uniform incorporation into the hydrogel precursor.

Having established stable LM@TA dispersions, we next incorporated them into a semi-interpenetrating polymer network (semi-IPN) to evaluate whether the TA coating could simultaneously fulfill its second function: establishing interfacial hydrogen bonds with the polymer matrix. The PHEA-S/LM@TA hydrogel was fabricated through a two-step process (Figure 1B). First, LM@TA nanodroplets were dispersed in an aqueous precursor containing hydroxyethyl acrylate (HEA), methyltrimethoxysilane (MTMS), urea, and cetyltrimethylammonium bromide (CTAB), followed by UV irradiation in the presence of Irgacure 2959. Prior studies have shown that ligand-stabilized LM droplets can initiate free-radical polymerization.⁴² In our system, both LM@TA and Irgacure 2959 contributed to initiating polymerization, whereas Irgacure-mediated UV irradiation was used to

ensure reliable and complete formation of a physically cross-linked PHEA network. Subsequent thermal-treatment-induced MTMS hydrolysis and condensation generated a covalent siloxane network that interpenetrated with the PHEA chains to form PHEA-silicone (PHEA-S) network. The siloxane covalent network serves as an elastic scaffold, imparting structural stability and strength. Urea accelerated MTMS hydrolysis while CTAB served as an interfacial compatibilizer to suppress phase separation.^{43,44} Importantly, the hydroxyl groups on PHEA chains not only formed interchain hydrogen bonds but also partially condensed with silanol species to create covalent Si–O–C linkages, while the phenolic hydroxyl groups on LM@TA were designed to form extensive hydrogen bonds with both networks.⁴⁵

Based on the above interfacial and network interactions, a schematic framework is proposed to elucidate how distinct filler-matrix coupling states regulate the crack-tip response of the hydrogel (Figure 1C). For rigid inclusions, the pronounced mechanical mismatch with the soft matrix induces interfacial debonding under deformation, creating localized gaps around the filler. The surrounding polymer chains are subsequently pulled out and stretched, leading to severe stress concentration at the crack tip, followed by chain scission and accelerated crack propagation. In the case of bare LM, although the intrinsically deformable droplets can partially relax local stress, the weak filler-matrix coupling results in interfacial decoupling and slippage between the droplets and polymer chains, limiting effective stress transfer and energy dissipation. By contrast, for LM@TA, catechol-mediated interfacial interactions establish robust yet dynamic coupling with the PHEA-S networks, enabling coordinated deformation, stress delocalization, and enhanced energy dissipation, thereby suppressing crack propagation.

To verify the structural basis for the proposed crack-regulation mechanism, FTIR spectroscopy was first conducted to confirm the successful formation of the multi-crosslinked architecture (Figure 2A). The disappearance of the vinyl peak at 1,633 cm^{−1} indicated complete polymerization of HEA monomers.³⁰ Meanwhile, the characteristic C–O stretching vibration of HEA at 1,081 cm^{−1} and the Si–O–C vibration of MTMS at 1,076 cm^{−1} merged into a broadened band centered at 1,071 cm^{−1}, accompanied by the emergence of a strong Si–O–Si absorption at 1,020 cm^{−1}, confirming both the formation of the siloxane network and the covalent bonding between PHEA and siloxane components.^{46–48} Most importantly, the –OH stretching vibration exhibited a progressive redshift from 3,422 cm^{−1} in HEA monomer to 3,412 cm^{−1} in PHEA-S and further to 3,380 cm^{−1} in PHEA-S/LM@TA, reflecting the enhanced hydrogen bonding interactions upon LM@TA incorporation.³⁰ XPS analysis provided additional evidence for the interfacial hydrogen bonding between LM@TA and the polymer matrix. The survey spectrum of PHEA-S/LM@TA showed a Ga 3d signal, confirming successful incorporation of LM@TA into the hydrogel (Figure 2B). High-resolution C 1s spectra (Figure 2C) revealed that the binding energy of the C=O shifted from 288.76 eV in PHEA-S to 288.88 eV in PHEA-S/LM@TA. A similar trend was observed in the O 1s spectra (Figure 2D), where the Si–O–Si/Si–OH shifted from 532.12 to 532.21 eV and the C–O moved from 533.34 to 533.54 eV.⁴⁶ These positive shifts indicate electron-density redistribution associated with hydrogen-bond formation

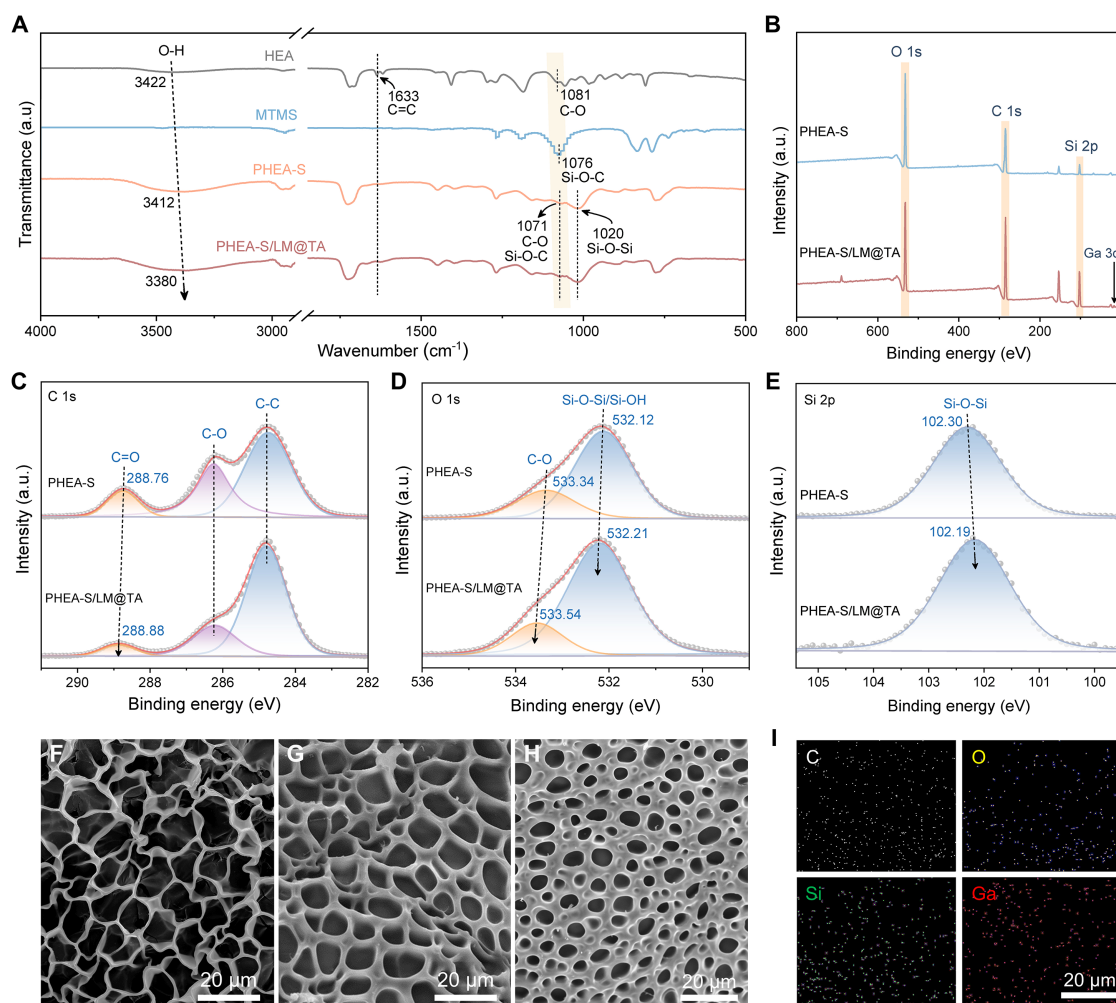


Figure 2. Structural characterization of the PHEA-S/LM@TA hydrogel

(A) FTIR spectra of the HEA monomer, MTMS monomer, PHEA-S and PHEA-S/LM@TA hydrogels, confirming polymerization, siloxane network formation, and enhanced hydrogen bonding interactions.

(B) XPS survey spectra of the PHEA-S and PHEA-S/LM@TA hydrogel, showing the appearance of Ga 3d signal after LM@TA incorporation.

(C–E) High-resolution (C) C 1s, (D) O 1s, and (E) Si 2p spectra, revealing binding energy shifts indicative of interfacial electronic interaction and hydrogen-bond formation.

(F–H) Cross-sectional SEM images of (F) PHEA, (G) PHEA-S, and (H) PHEA-S/LM@TA hydrogels, respectively, showing progressive pore densification and structural homogenization (scale bars, 20 μm).

(I) EDS elemental mapping, showing uniform LM@TA (Ga) distribution in the hydrogel matrix (scale bars, 20 μm).

between the phenolic hydroxyl groups of LM@TA and the oxygen-containing moieties in the polymer network. In addition, the Si 2p signal exhibited a slight shift from 102.30 to 102.19 eV (Figure 2E),⁴⁹ further suggesting that the local siloxane chemical environment was also perturbed by LM@TA incorporation. Together, these results indicate that LM@TA was integrated into the PHEA-S network through strong hydrogen bonds rather than simple physical entrapment. Density functional theory (DFT) calculations were performed using Ga₂O₃ as a model surface for the oxidized LM interface to elucidate the interfacial role of TA-modified LM.³¹ As shown in Figure S7A, the adsorption energy of the PHEA-S/LM@TA model (−1.52 eV) is lower than that of the PHEA-S/LM model (−1.14 eV), indicating enhanced interfa-

cial affinity after TA modification.^{50,51} Charge density difference analysis revealed more pronounced charge redistribution at both the TA/Ga₂O₃ and TA/PHEA-S interfaces in the PHEA-S/LM@TA model (Figure S7B).⁵² These results confirm that the TA-mediated interface establishes strong interfacial affinity of the LM@TA with the PHEA-S network. Scanning electron microscopy (SEM) further revealed the structural consequence of this interfacial integration. Compared with the relatively looser porous morphologies of pure PHEA and PHEA-S hydrogels, the PHEA-S/LM@TA hydrogel exhibited a denser porous architecture (Figures 2F–2H). The corresponding EDS elemental mapping of PHEA-S/LM@TA hydrogel confirmed the uniform distribution of C, O, Si, and Ga throughout the matrix (Figure 2I).

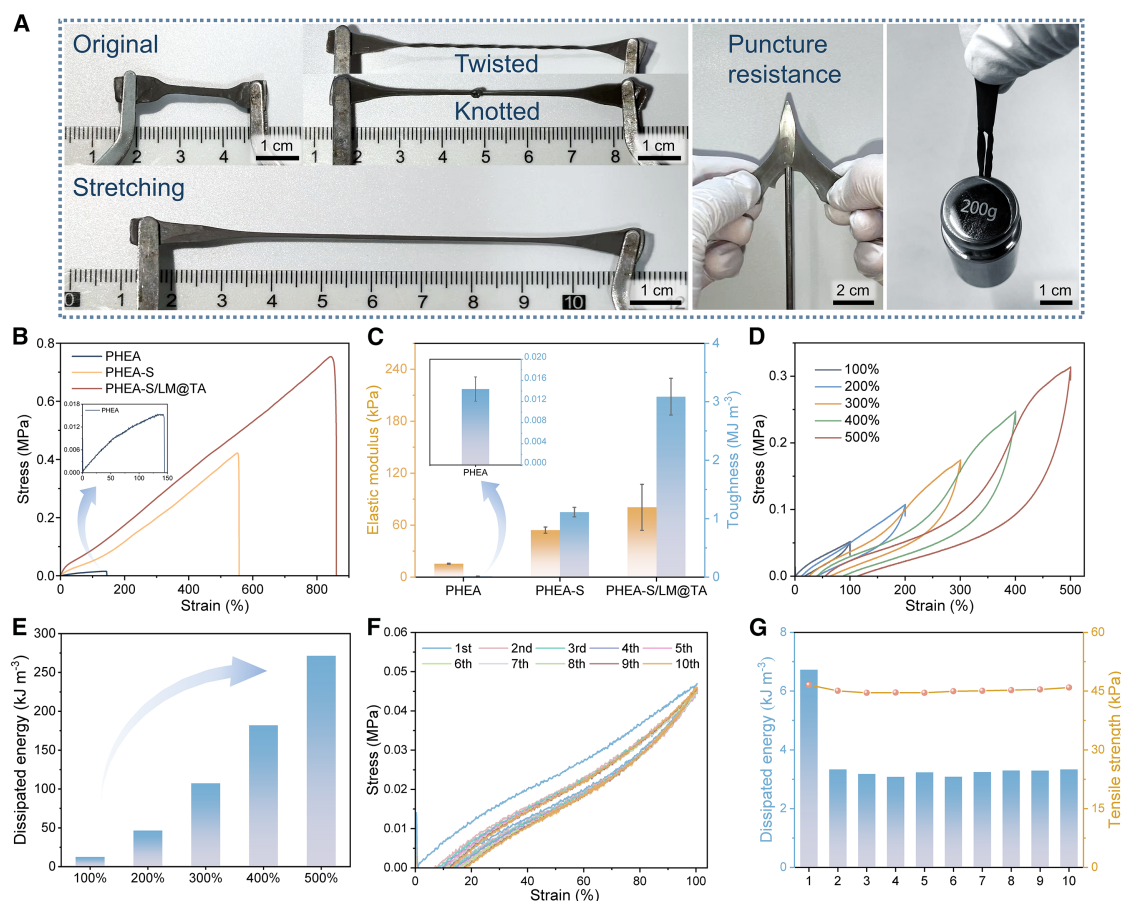


Figure 3. Tensile mechanical characterization of the PHEA-S/LM@TA hydrogel

(A) Photographs of the PHEA-S/LM@TA hydrogel samples showing structural integrity under stretching, twisting, knotting, puncturing, and lifting a 200 g weight, illustrating excellent mechanical performances (scale bars, 1 cm).

(B and C) Comparison of (B) tensile stress-strain curves and (C) the corresponding elastic modulus and toughness of the PHEA, PHEA-S, and PHEA-S/LM@TA hydrogels, highlighting the progressive mechanical reinforcement. Data are represented as mean $\pm$ SD ($n = 3$).

(D and E) (D) Cyclic loading-unloading tensile curves and (E) the corresponding dissipated energies of the PHEA-S/LM@TA hydrogel at different maximum strains (100%, 200%, 300%, 400%, and 500%), showing the remarkable energy dissipation.

(F and G) (F) Ten successive cyclic tensile loading-unloading curves and (G) the corresponding dissipated energies and tensile strengths of the PHEA-S/LM@TA hydrogel under 100% strain, demonstrating excellent cyclic stability.

These results demonstrate that TA coating fulfills its second key function: establishing abundant interfacial hydrogen bonds between LM@TA nanodroplets and the polymer network, thereby achieving strong filler-matrix integration that is essential for efficient stress transfer and energy dissipation.

Enhanced stretchability and toughness through multi-scale hydrogen bond dissipation

The unique multi-crosslinked architecture, together with robust hydrogen bonding and uniform dispersion of LM@TA within the polymer matrix, markedly enhanced the mechanical properties of the PHEA-S/LM@TA hydrogel. As shown in Figure 3A, the hydrogel samples sustained large uniaxial stretching, twisting, and knotting, puncturing and even lifting a 200 g weight without structural failure, demonstrating exceptional stretchability, strength, and toughness. Quantitative tensile tests were used

to reveal the progressive mechanical enhancement achieved through semi-IPN formation and LM@TA incorporation (Figure 3B). The pure PHEA hydrogel showed limited mechanical properties, with a fracture stress of 0.015 MPa and fracture strain of 144%. After forming a semi-interpenetrating network with silicone, the fracture stress and fracture strain of the PHEA-S hydrogel increased to 0.42 MPa and 553%, respectively. Incorporating LM@TA further boosted mechanical performance, yielding a fracture stress of 0.75 MPa and fracture strain of 860%. Correspondingly, both the elastic modulus and toughness were significantly improved (Figure 3C). In particular, the PHEA-S/LM@TA hydrogel achieved a high toughness of 3.09 MJ m^{-3} , approximately 2.8 times that of the PHEA-S hydrogel (1.11 MJ m^{-3}). The reinforcing effect of LM@TA nanodroplets can be discussed from the perspective of elastocapillary mechanics.⁵³ Unlike simple liquid inclusions, oxide-coated LM droplets possess an

interfacial layer with viscoelastic and yielding behavior.⁵⁴ The characteristic elastocapillary length is estimated using the effective interfacial stress (often represented experimentally by the surface yield stress of the oxide shell) for oxide-coated LM droplets.⁵⁵ Although the surrounding aqueous environment and ligand coordination may affect the surface yield stress of oxide-coated LM,^{56–58} the effective interfacial stress of TA-functionalized LM droplets has not yet been experimentally determined. Therefore, the reported surface yield stress of native oxide-coated LM ($0.2\text{--}0.5\text{ N m}^{-1}$) was adopted as an order-of-magnitude estimate of the interfacial resistance.⁵⁹ Together with the elastic modulus of the PHEA-S matrix (54.2 kPa), the characteristic elastocapillary length was estimated to be approximately $3.7\text{--}9.2\text{ }\mu\text{m}$. Since the LM@TA droplet radius ($\sim 0.2\text{ }\mu\text{m}$) is substantially smaller than this estimate, interfacial mechanics are expected to strongly constrain droplet deformation at small strain. This analysis is consistent with, but does not by itself prove, a stiffening contribution from LM@TA. Although the free liquid-like deformation of LM@TA nanodroplets is expected to be restricted under small strain, the strong TA-mediated interfacial coupling enables constrained cooperative deformation of the nanodroplets together with the surrounding polymer network under large deformation.⁶⁰ More importantly, the multi-scale hydrogen bond dissipation within the PHEA-S/LM@TA network largely contributes to pronounced improvements of the mechanical performance. Covalent and hydrogen bonds between PHEA chains and the covalent silicone network provide inter-network connectivity, bearing part of the load and facilitating efficient energy dissipation,⁴⁷ which was further verified by the fact that PHEA/LM@TA hydrogel without silicone network exhibited weak mechanical performances (Figure S8A). Moreover, LM@TA functions as a multifunctional crosslinker, forming abundant hydrogen bonds with the polymer matrix.^{45,61} During deformation, these hydrogen bonds can largely dissipate energy through reversible rupture, while the deformable LM@TA nanodroplets enable efficient stress transfer among polymer chains, collectively contributing to the superior mechanical performance of the PHEA-S/LM@TA hydrogel.³⁰ This hypothesis is supported by the PHEA-S/LM (without TA-mediated dynamic interface) hydrogel exhibiting a lower fracture strain (623%) and fracture stress (0.30 MPa) due to the weak filler-matrix interactions (Figure S8B).

The multi-crosslinked architecture of the hydrogel can be tuned by adjusting LM@TA content, PHEA and silicone compositions, and solid content, thereby modulating its mechanical properties. The influence of LM@TA content on the mechanical properties of the PHEA-S/LM@TA hydrogel was investigated (Figure S9), revealing that fracture stress, fracture strain, elastic modulus, and toughness initially increased and then decreased with LM@TA content, reaching an optimum at 3.3 wt %. The incorporation of LM@TA up to this optimal level enhanced the crosslinking density, whereas excessive LM@TA ($>3.3\text{ wt }%$) may induce structural defects due to LM@TA agglomeration, disrupting network continuity and deteriorating tensile properties.²¹ The HEA:MTMS molar ratio also critically regulated mechanical behavior (Figure S10), with a ratio of 3:2 yielding the highest fracture stress, fracture strain, and toughness due to the synergistic interaction between the physically crosslinked

PHEA network and the covalently crosslinked siloxane network, where the covalent siloxane serves as elastic skeleton and the physical interactions act as effective energy-dissipation units.⁶² Furthermore, increasing the solid content markedly improved mechanical strength, with performance plateauing above 50 wt % (Figure S11). Based on these optimization studies, the PHEA-S/LM@TA hydrogel formulated with 50 wt % solid content, a HEA:MTMS molar ratio of 3:2, and 3.3 wt % LM@TA exhibited an excellent combination of fracture stress (0.75 MPa), fracture strain (860%), elastic modulus (80.62 kPa), and toughness (3.09 MJ m^{-3}), and was therefore selected for all subsequent studies.

The multi-scale hydrogen-bonding network in the PHEA-S/LM@TA hydrogel can reversibly dissociate and reform, enabling efficient energy dissipation and cyclic stability. As shown in Figures 3D and 3E, successive loading-unloading tests conducted at increasing maximum strains generated progressively enlarged hysteresis loops, with the dissipated energy rising from 12.69 kJ m^{-3} at 100% strain to 271.61 kJ m^{-3} at 500%. These results indicate that under larger strain, more hydrogen bonds between the PHEA chains, silicone network and LM@TA were effectively dissociated to dissipate more energy, contributing to high toughness.⁶³ A comparison of the energy dissipation behaviors of PHEA-S and PHEA-S/LM@TA hydrogels further highlights the role of the covalent siloxane network as an elastic framework, as well as the critical contribution of LM@TA-polymer dynamic hydrogen bonding to the enhanced toughness of the composite system (Figure S12). Additionally, 10 consecutive loading-unloading cycles performed at 100% strain showed that a pronounced hysteresis loop was observed with dissipated energy of 6.73 kJ m^{-3} in the first cycle (Figures 3F and 3G), further indicating the rapid dissociation of abundant hydrogen bonds in PHEA-S/LM@TA to dissipate energy and protect the integrity of the hydrogel network. In subsequent cycles, the hysteresis loops almost completely overlapped with the dissipated energy stabilizing at 3.23 kJ m^{-3} , reflecting the rapid reconstruction of the partially ruptured hydrogen bonds. Notably, the tensile stress in the tenth cycle (45.91 kPa) remained approximately 98.6% of that in the first cycle (46.56 kPa), demonstrating remarkable cyclic durability and self-recovery capability.

Coordinated soft filler-matrix deformation for excellent flaw insensitivity

The crack-resistant capability is a crucial property that enables materials to withstand mechanical damage, thereby extending their service lifetime.⁶⁴ The soft LM@TA-composited multi-crosslinked structure endows the PHEA-S/LM@TA hydrogel with exceptional crack resistance and fracture toughness. Fracture resistance of the hydrogel was evaluated by determining the fracture energy (Γ) using the Rivlin-Thomas single-edge notch method.⁶⁵ For PHEA-S hydrogel, introducing a notch ($a/w = 0.4$) reduced fracture strain from 600% to 328%, with a low fracture energy of 2.6 kJ m^{-2} (Figure 4A; Figure S13B), indicating poor crack resistance. Under the same notched condition, the PHEA-S/LM hydrogel exhibited a fracture energy of only 2.0 kJ m^{-2} , even lower than that of PHEA-S (Figures S13A and S13B). In contrast, the PHEA-S/LM@TA hydrogel exhibited a fracture strain of 724% under identical notched conditions,

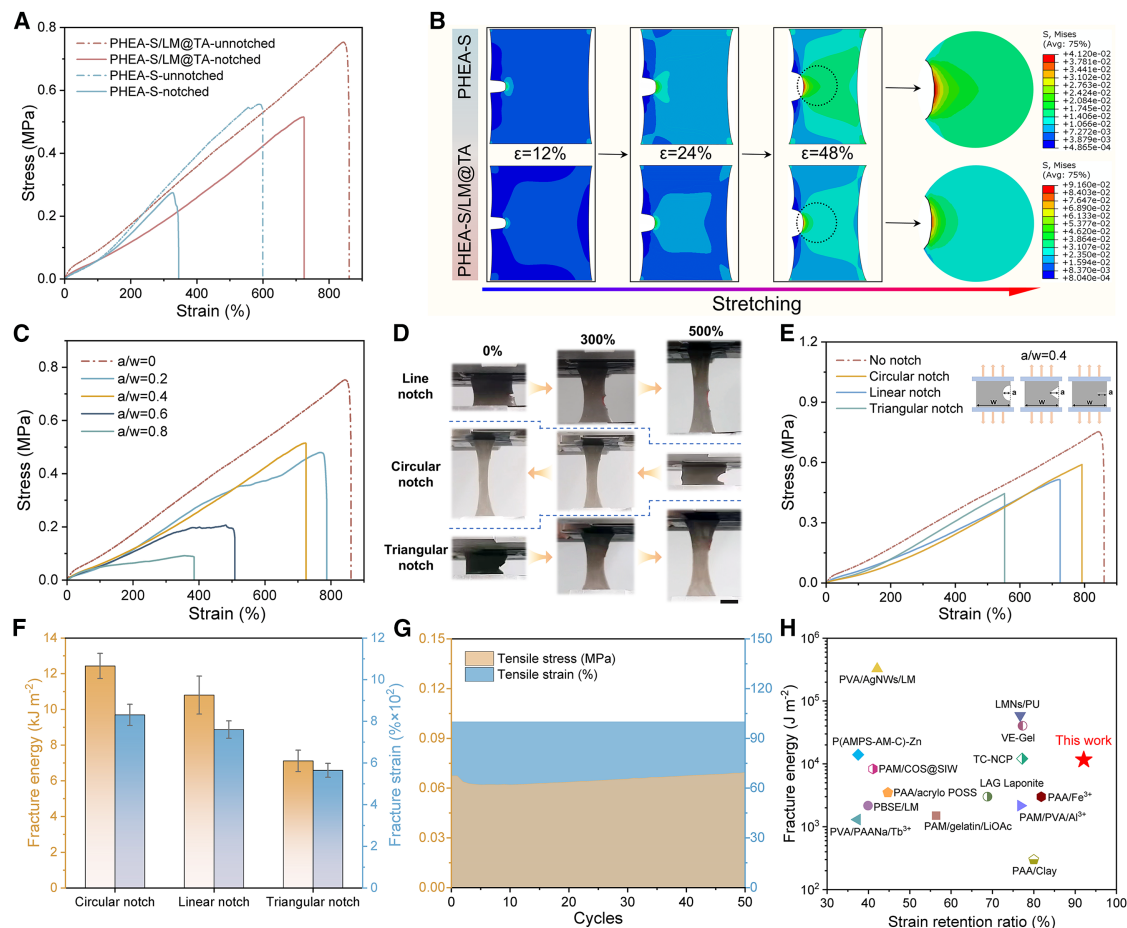


Figure 4. Flaw insensitivity and fatigue resistance of the PHEA-S/LM@TA hydrogel

(A) Comparison of tensile stress-strain curves of unnotched and single-edge notched ($a/w = 0.4$) PHEA-S and PHEA-S/LM@TA hydrogels.
 (B) Finite element analysis (FEA) of stress distribution in notched hydrogels under tensile strain, showing stress delocalization in the PHEA-S/LM@TA hydrogel compared to the PHEA-S hydrogel.
 (C) Tensile stress-strain curves of the PHEA-S/LM@TA hydrogel with linear notches of varying lengths ($a/w = 0-0.8$).
 (D) Optical photographs of PHEA-S/LM@TA hydrogel samples with circular, linear, and triangular notches stretched perpendicular to the notch direction, demonstrating minimal crack propagation (scale bar, 2 cm).
 (E and F) Tensile stress-strain curves and (F) the corresponding fracture energies and fracture strains of PHEA-S/LM@TA hydrogels with various notch shapes. Data are represented as mean $\pm$ SD ($n = 3$).
 (G) Cyclic tensile stress curves of the notched PHEA-S/LM@TA hydrogel over 50 cycles at 100% strain.
 (H) Comparison of fracture energy and strain retention ratio of the PHEA-S/LM@TA hydrogel with previously reported nanocomposite hydrogels.

with a strain retention ratio of 84%. Moreover, its fracture energy reached 10.8 kJ m^{-2} , approximately four times higher than that of PHEA-S hydrogel. Optical microscopy images (Figure S14) showed pronounced aggregation of bare LM within the PHEA-S/LM hydrogel, whereas TA-modified LM@TA droplets were uniformly dispersed throughout the PHEA-S/LM@TA matrix. These results suggest that, without interfacial modification, the bare LM cannot effectively blunt crack-tip stress and instead facilitates premature failure at the defect region due to the local aggregation of bare LM, together with its weak interfacial coupling with the hydrogel matrix. Optical microscopy observations of the PHEA-S/LM@TA hydrogel further reveal that the initially randomly distributed LM@TA domains progressively aligned along the loading direction upon stretching to 100% and 200%

strain; upon further increasing the strain to 400%, these domains underwent pronounced elongation and evolved into a more continuous and highly oriented morphology (Figure S15). These observations indicate that the LM@TA nanodroplets deform cooperatively with the PHEA-S network during tensile loading owing to the strong interfacial coupling between the soft LM@TA nanodroplets and the hydrogel matrix. Consistently, no LM leakage was observed during the deformation of notched PHEA-S/LM@TA hydrogels, further confirming the robust confinement and effective interfacial integration of LM@TA within the polymer network (Figure S16 and Video S1). Meanwhile, the reversible rupture and reformation of interfacial hydrogen bonds enable the LM@TA nanodroplets to serve as effective stress-transfer and energy-dissipation centers, thereby

facilitating stress redistribution and alleviating crack-tip stress concentration. This multi-level energy dissipation mechanism facilitated by soft LM@TA and collaborative deformation contributed to the remarkable enhancement in crack resistance. Finite element analysis (FEA) of notched hydrogels under tensile strain (Figure 4B) revealed a broad, smooth stress distribution in the PHEA-S/LM@TA hydrogel, contrasting with localized stress in the PHEA-S hydrogel, demonstrating effective stress delocalization that mitigates localized failure and enhances fracture toughness.

The flaw insensitivity of the PHEA-S/LM@TA hydrogel was further evaluated by using single-notch tests with varying notch depths. As the normalized notch length (a/w) increased from 0 to 0.8, fracture strain decreased from 860% to 385%, while fracture energy exhibited a non-monotonic trend, increasing from 2.8 kJ m⁻² (a/w = 0.2) to 11.9 kJ m⁻² (a/w = 0.6) and slightly decreasing at a/w = 0.8, yet remaining 9.0 kJ m⁻², substantially higher than PHEA-S hydrogel (Figure 4C; Figure S17), indicating excellent flaw tolerance. The PHEA-S/LM@TA hydrogel also displayed notch-shape insensitivity: circular, linear, and triangular-notched samples endured high strains under perpendicular loading with minimal crack propagation (Figure 4D and Videos S2, S3, and S4). Fracture strains of circular-, linear-, and triangular-notched samples reached 792% (strain retention ratio of 92%), 760% (strain retention ratio of 88%), and 559% (strain retention ratio of 65%), with corresponding fracture energies of 12.43, 10.80, and 7.12 kJ m⁻² (Figures 4E and 4F). Additionally, fatigue resistance was assessed via 50-cycle tensile tests at 100% strain, showing negligible crack propagation and minimal stress decay (Figure 4G; Figure S18). Tear resistance evaluated by trouser tear tests (Figure S19) showed the tearing energy of PHEA-S/LM@TA hydrogel was as high as 4.52 kJ m⁻², significantly surpassing that of the PHEA-S hydrogel (0.18 kJ m⁻²). Compared with reported LM-based and other nanocomposite hydrogels, PHEA-S/LM@TA hydrogel combined high fracture toughness with superior strain retention ratio (Figure 4H; Table S3), underscoring its potential for flexible wearable devices under extreme mechanical conditions.

Photothermal functionality from plasmonic resonance of LM nanodroplets

The PHEA-S/LM@TA hydrogel exhibited highly efficient photothermal conversion performance, mainly attributed to the localized surface plasmon resonance (LSPR) of EGaIn nanodroplets under near-infrared (NIR) irradiation, where the optical field induces collective oscillations of free electrons to achieve rapid light-to-heat conversion (Figure 5A).^{66,67} Under 808 nm irradiation (0.05 W cm⁻²), the surface temperature of the PHEA-S/LM@TA hydrogel increased sharply from 23.7°C to 57.8°C within 40 s and quickly returned to baseline after the irradiation stopped (Figures 5B and 5C). In contrast, the LM-free PHEA-S hydrogel showed a much slower heating rate and a significantly lower maximum temperature, confirming that LM@TA dramatically enhances photothermal efficiency. The photothermal response was further modulated by varying the laser power, with higher power densities yielding faster heating and elevated steady-state temperatures up to 84.5°C at 2.0 W cm⁻² (Figures S20A and S20B). The PHEA-S/LM@TA hydrogel also demonstrated

excellent photothermal stability, maintaining nearly identical heating and cooling profiles over five on-off irradiation cycles (Figure 5D). In practical photothermal applications, materials inevitably suffer from mechanical damage, thus maintaining mechanical robustness under photothermal operating conditions is critical for photothermal devices. Accordingly, we evaluated notched PHEA-S/LM@TA hydrogels under NIR-induced heating. As shown in Figure S21 and Video S5, even with a pre-cut notch and heating to 43.7°C, the hydrogel retained a tensile strength of 0.42 MPa, a fracture strain of 697%, and a fracture energy of 9.53 kJ m⁻². The preservation of flaw insensitivity under thermal activation is consistent with the dynamic nature of the poly-phenol-mediated interface, where mild heating accelerates hydrogen-bond exchange without compromising stress transfer. The hydrogel therefore retains robust notch tolerance throughout the photothermal working window.

These rapid, reversible, and power-dependent photothermal characteristics enable real-time infrared camouflage. When applied to the surface of a toy duck, the PHEA-S/LM@TA hydrogel rapidly adjusted its surface temperature to match environmental backgrounds ranging from 25°C to 65°C, achieving dynamic thermal concealment by simply tuning irradiation intensity and duration (Figure 5E). Moreover, when wrapped around a finger (Figure 5F), the PHEA-S/LM@TA hydrogel effectively suppressed the infrared emission of the underlying heat source, rendering it nearly imperceptible to thermal imaging cameras, whereas the PHEA-S hydrogel provided negligible shielding. Together, these results highlight the strong potential of PHEA-S/LM@TA hydrogels for concealing body-heat signatures and protecting flexible devices in defense-related applications.

Damage-tolerant strain sensing and electrophysiological signal acquisition

The incorporation of LM@TA nanodroplets endows the PHEA-S/LM@TA hydrogel with good electrical conductivity. As the LM@TA content increased to 9.9 wt %, the conductivity of the hydrogel increased from 0.09 S · m⁻¹ (without LM@TA) to 0.47 S · m⁻¹, as further evidenced by the successful illumination of an LED (Figure S22). The good conductivity originates from the transport and hopping of electrons in LM@TA and the migration of free ions (such as ions ionized from CTAB and water) via free water.⁶⁸ Although with TA insulating layer, effective LM-dominated electronic network can be formed through near-contact percolation and electron tunneling between adjacent LM cores; meanwhile, the aqueous phase provides a parallel ionic conduction and alleviates interfacial polarization, together ensuring good conductivity.⁶⁹ The strain-sensing performance was evaluated by monitoring the relative resistance change ($\Delta R/R_0$) under applied strain (ϵ), with sensitivity quantified by the gauge factor ($GF = \Delta R/R_0/\epsilon$).⁷⁰ As shown in Figure 6A, the hydrogel exhibited distinct linear sensing regimes, with GF values of 1.52 for 0%–250% strain and 3.55 for 250%–860% strain, indicating high sensitivity over a wide strain range. This sensitivity change arises from the strain-dependent evolution of the conductive network. At low strain, LM nanoparticles remain closely packed, forming continuous and stable conductive pathways. As strain increases, particle separation reduces effective contact interfaces, elevating contact resistance. In the high-strain regime,

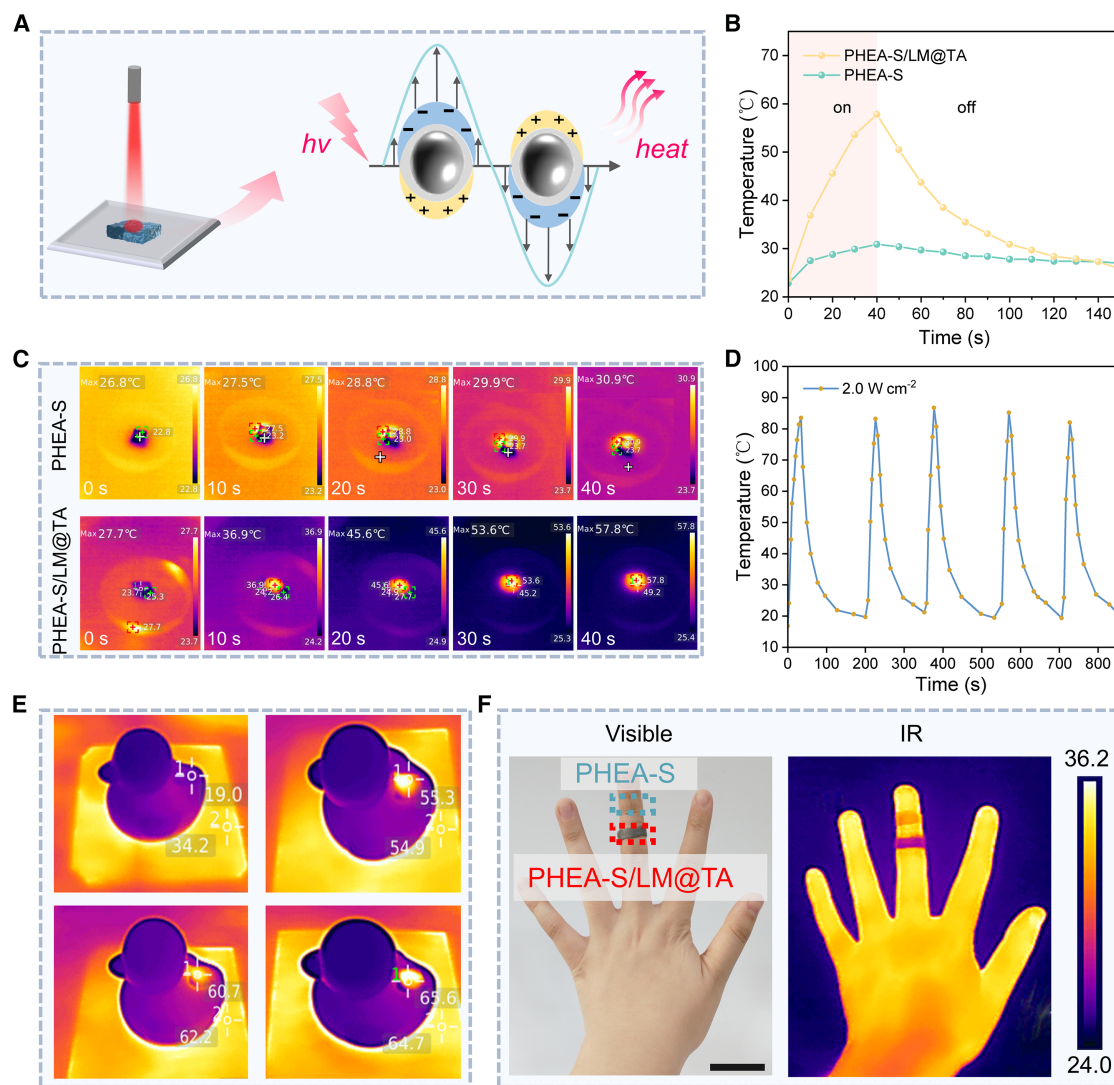


Figure 5. Photothermal conversion behavior and infrared camouflage performance of the PHEA-S/LM@TA hydrogel

(A) Schematic illustration of the photothermal conversion mechanism of LM nanodroplets under NIR irradiation via localized surface plasmon resonance (LSPR). (B and C) Real-time temperature profiles and (C) corresponding infrared thermal images of PHEA-S and PHEA-S/LM@TA hydrogels under 808 nm NIR irradiation (0.05 W cm^{-2}), demonstrating markedly enhanced photothermal efficiency after LM@TA incorporation.

(D) Photothermal stability of the PHEA-S/LM@TA hydrogel over repeated on/off irradiation cycles.

(E) Dynamic infrared camouflage on a toy surface enabled by rapid temperature modulation of the PHEA-S/LM@TA hydrogel.

(F) Visible and infrared images of PHEA-S and PHEA-S/LM@TA hydrogels wrapped around a human hand, highlighting the superior infrared shielding capability (scale bar, 2 cm).

rapid disruption of interparticle connections limits charge-transport pathways, causing a sharp resistance increase and a corresponding enhancement in sensitivity.⁹ Figure 6B shows that $\Delta R/R_0$ increased steadily with strain up to 90%, fully recovering upon release, and maintained stable output during constant strain-holding stages, demonstrating reliable real-time multi-cycle monitoring. Instantaneous stretching-releasing tests revealed a response time of 113 ms and recovery time of 142 ms (Figure S23A). Across micro-strain (3%–9%) (Figure 6C), moderate strain (10%–40%) (Figure 6E), and large strain (50%–200%) (Figure S23B) cycles, the $\Delta R/R_0$ signal demonstrated clear dif-

ferentiation between strain levels and high repeatability at identical strains. The hydrogel also exhibited rate-independent sensing characteristics with negligible variation of $\Delta R/R_0$ at the fixed strain of 200% increasing stretching rate (Figure S23C). Furthermore, the hydrogel sensor retained stable performance over 300 loading-unloading cycles at 50% strain (Figure 6D), underscoring its excellent durability.

The outstanding crack resistance of the PHEA-S/LM@TA hydrogel ensures that sensor functionality is preserved even under partial structural damage. The GF of notched PHEA-S/LM@TA hydrogel was first assessed (Figure S24). The notched sensor

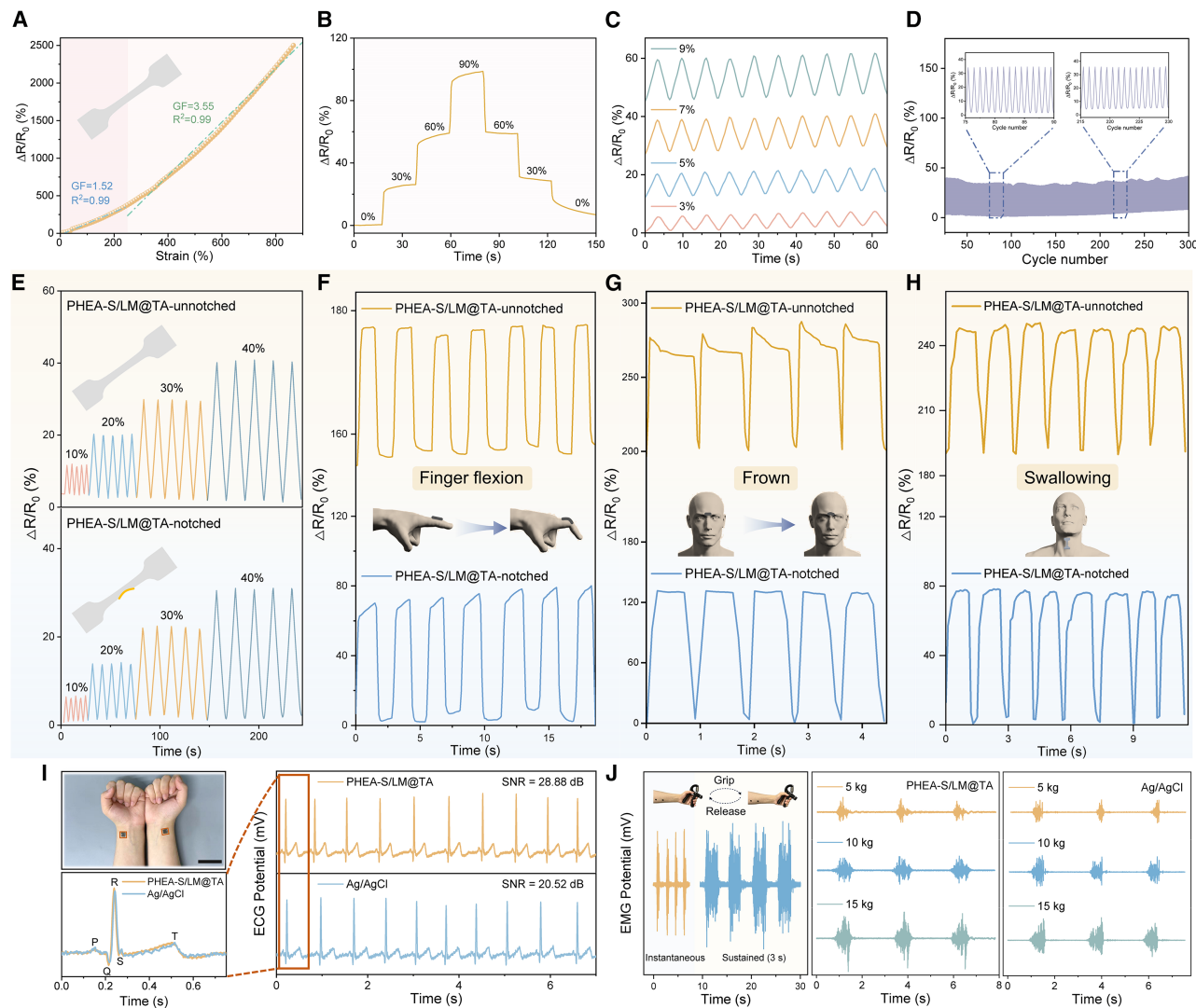


Figure 6. Damage-tolerant strain sensing and bioelectronic applications of the PHEA-S/LM@TA hydrogel

(A) Relative resistance change ($\Delta R/R_0$) versus strain and corresponding gauge factor (GF) of the intact hydrogel sensor, with GFs of 1.52 for 0%–250% strain and 3.55 for 250%–860% strain.

(B) $\Delta R/R_0$ during progressive stretching and releasing up to 90% strain.

(C) Cyclic $\Delta R/R_0$ response at slight strain (3%–9%).

(D) Stability over 300 cyclic loading-unloading cycles at 50% strain.

(E) $\Delta R/R_0$ of intact and notched hydrogels under cyclic small strains (10%–40%).

(F–H) Sensor responses of intact and notched hydrogels during dynamic motions: finger flexion (F), frowning (G), and swallowing (H).

(I) ECG signals recorded by the hydrogel electrode compared with a commercial Ag/AgCl electrode (scale bar, 5 cm).

(J) EMG signals captured by the hydrogel electrode during rapid and sustained grip-release cycles under varying forces (5, 10, and 15 kg), compared with Ag/AgCl electrode.

exhibited well-defined piecewise-linear sensing behavior, with GF values of 1.01 (0%–300%), 1.91 (300%–550%), and 3.51 (550%–724%), respectively. Notably, these GF values were comparable to those of the intact sensor, and the characteristic strain-dependent evolution of GF was well preserved, indicating that the deformation-induced modulation of the conductive network remains effective despite the presence of a notch. Moreover, the notched hydrogel sample exhibited stable and repeatable responses under cyclic loading-unloading at different

small strains (10%, 20%, 30%, and 40%), consistent with intact performance (Figure 6E). The notched sample also maintained reliable signals under large periodic strains (50%, 100%, and 150%; Figure S25A). Furthermore, similar to the behavior of the intact sensor, the notched sensor exhibited highly consistent resistance responses across all tested strain rates (Figure S25B), indicating negligible rate dependence. During 300 consecutive tensile-release cycles at 10% strain, the $\Delta R/R_0$ response of notched hydrogels showed negligible attenuation or drift

(Figure S25C), demonstrating excellent fatigue resistance and structural integrity. These results indicate that localized damage neither propagates under cyclic loading nor compromises sensor performance, highlighting the hydrogel's defect tolerance and suitability for reliable flexible sensing under demanding conditions.

Leveraging its superior sensing performance and exceptional damage tolerance, both unnotched and notched PHEA-S/LM@TA hydrogels were assembled into wearable sensors for human motion monitoring. As shown in Figures 6F–6H, the response signals of notched sensors closely matched those of intact sensors during finger bending, frowning, and swallowing. Despite the mechanical defects, notched sensors faithfully reproduced all key features of the intact signals, accurately tracking sustained finger flexion-relaxation, resolving subtle facial muscle movements, and capturing transient swallowing events. The only difference was a constant amplitude offset due to initial resistance variations, with no noticeable differences in waveform evolution or response speed. The robustness of the notched sensors was further confirmed under large-deformation scenarios, where cyclic bending of the wrist, elbow, and knee produced stable and repeatable responses (Figures S26A–S26C). Furthermore, reliable and repeatable electrical signals were obtained during finger-bending monitoring after NIR irradiation (Figure S27), confirming that the sensing accuracy and signal stability can be maintained even under the combined challenges of photothermal activation and structural damage. Collectively, the PHEA-S/LM@TA hydrogel combines high sensitivity, rapid response, excellent cyclic stability, and a wide strain detection range with exceptional notch insensitivity, enabling adaptability to complex service conditions and positioning it as a promising platform for next-generation damage-tolerant and durable flexible electronic devices.

To evaluate practical bioelectronic applications, the PHEA-S/LM@TA hydrogel was employed as a skin-mounted bioelectrode for recording electrophysiological signals, including electrocardiograms (ECG) and electromyograms (EMG). Due to their skin-like flexibility and efficient ionic/electronic transport, PHEA-S/LM@TA hydrogel bioelectrodes exhibited lower interfacial impedance than commercial Ag/AgCl electrodes over the entire frequency range (10^{-1} – 10^5 Hz) (Figure S28), favoring the high-quality acquisition of bioelectrical signals. For ECG monitoring, the hydrogel electrode attached to the wrist captured well-defined PQRST waveforms with clearly distinguishable peaks, which were nearly the same as signals from a commercial Ag/AgCl electrode (Figure 6I). Quantitative analysis revealed that the ECG signal-to-noise ratio (SNR) of the hydrogel electrode reached 28.88 dB, exceeding the 20.52 dB of the Ag/AgCl reference, demonstrating superior signal quality. The hydrogel electrode also effectively monitored dynamic heart-rate changes, recording an increase from 74 bpm at rest to 126 bpm after exercise (Figure S29). Continuous monitoring over 400 s showed stable waveform morphology and a consistent baseline (Figure S30), confirming long-term operational reliability. The hydrogel electrodes also exhibited high-quality EMG detection. During handgrip exercises at 5, 10, and 15 kg, EMG signal amplitudes increased proportionally with grip force, accurately reflecting forearm muscle contractions (Figure 6J). Similarly, during

dumbbell lifting at 1, 1.5, and 2.5 kg, the electrode clearly distinguished varying levels of muscle activation (Figure S31). These results demonstrate that the PHEA-S/LM@TA hydrogel provides robust, high-fidelity bioelectrical signal acquisition, establishing a reliable platform for multifunctional wearable health-monitoring devices.

Closed-loop recovery of LM via network degradation

The PHEA-S/LM@TA hydrogel demonstrates intrinsic degradability that enables efficient recycling of LM, in contrast to conventional conductive nanofillers-based hydrogels that are typically non-recyclable. This recyclability arises from the dynamic hydrogen-bonding and siloxane networks within the polymer matrix. Under alkaline conditions, these dynamic linkages are cleaved, leading to dissociation of the crosslinked structure. Disruption of the interfacial stabilizing layer surrounding LM nanodroplets increases the surface tension, promoting aggregation and coalescence into bulk LM, thereby facilitating separation from the aqueous phase.³⁰ As shown in Figure 7Ai–iii, complete hydrogel degradation was achieved within 24 h in a 4 M NaOH solution, followed by washing and separating to recover LM. The recovered LM showed only minor changes in appearance relative to fresh LM, likely due to slight surface oxidation, while maintaining excellent fluidity (Figure S32A). The recovery rate of LM reached $78.5 \pm 3.6\%$ (Figure S32B), demonstrating its good recyclability. Moreover, traces patterned with recycled LM could readily light an LED when integrated into a closed circuit under an applied voltage. The sheet resistance of a recycled-LM trace (measuring $1.5 \text{ cm} \times 0.4 \text{ cm}$) was approximately 0.1Ω , comparable to that of a fresh-LM trace measured under identical conditions (Figure 7A), indicating that the recovered LM retained metallic conductivity. SEM imaging with elemental mapping (Figure 7B) confirmed preservation of spherical morphology and presence of C, O, Ga, and In, indicating partial retention of TA on the LM surface. This sustainable degradation-recovery strategy provides an effective route for environmentally responsible recycling of LM-based soft electronics.

DISCUSSION

In this study, we have developed a highly stretchable and tough LM-composited hydrogel with exceptional resistance to crack propagation by incorporating soft LM@TA nanodroplets into a semi-interpenetrating PHEA-siloxane network. The TA interfacial modification markedly improved the stability and uniform dispersion of LM@TA nanodroplets within the hydrogel matrix, while simultaneously strengthening their interfacial bonding with the polymer network via abundant hydrogen-bonding interactions. This multiscale energy dissipation, arising from reversible rupture of interfacial hydrogen bonds and coordinated deformation of LM@TA with polymer chains, effectively dissipated mechanical energy and alleviated stress concentration at crack tips. As a result, the PHEA-S/LM@TA hydrogel exhibited an exceptional combination of high extensibility (860%), high strength (0.75 MPa), remarkable toughness (3.09 MJ m^{-3}), and extraordinary flaw insensitivity (fracture strain retention of up to 92% and fracture energy of 12.43 kJ m^{-2}). Beyond its

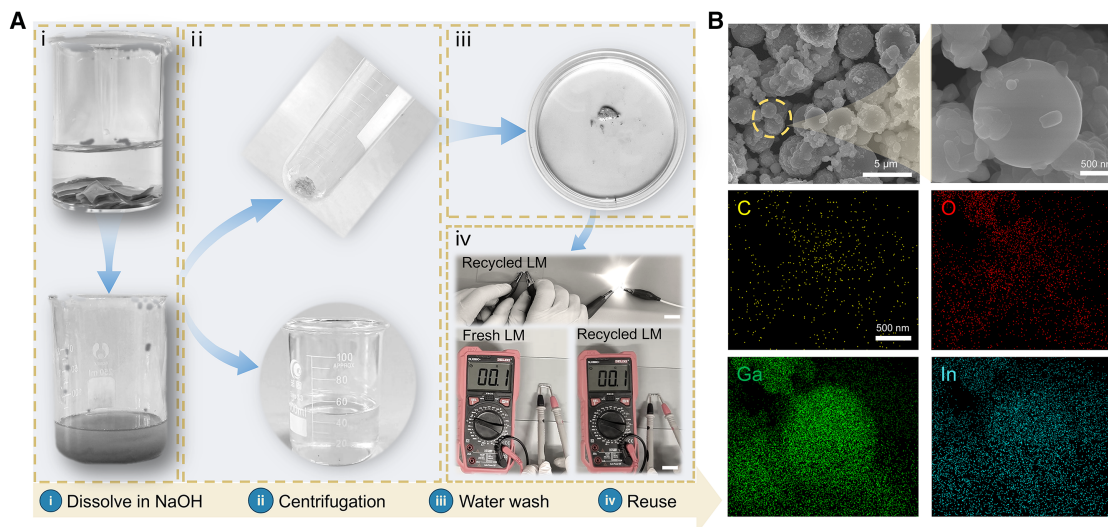


Figure 7. Closed-loop recovery of LM from the PHEA-S/LM@TA hydrogel

(A) Schematic of the recovery process via degradation of PHEA-S/LM@TA network: (i) dissolution in 4 M NaOH, (ii) separation by centrifugation, (iii) photograph of the recovered LM, and (iv) resistance measurement of the recycled and fresh LM (all samples were spread into rectangular strips), demonstrating retention of functional properties (scale bars, 2 cm).

(B) SEM image and corresponding elemental mapping of recovered LM droplets (scale bar, 5 μm), showing preserved spherical morphology and presence of C, O, Ga, and In, indicative of partial TA retention on the LM surface (scale bars, 500 nm).

mechanical robustness, the hydrogel demonstrated efficient photothermal conversion and adequate electrical conductivity. Hydrogel-based sensors showed high sensitivity over a broad working range, superior durability, and reliable performance even under mechanical damage. Consequently, wearable devices fabricated from this hydrogel enabled accurate real-time monitoring of diverse human motions and electrophysiological signals, while also exhibiting strong potential for infrared camouflage applications. This work establishes polyphenol-mediated interfacial engineering as a concrete design principle for damage-tolerant LM-composited hydrogels, with anticipated extension to broader soft-filler chemistries for soft robotics, wearable electronics, and bio-integrated devices.

METHODS

Materials

Eutectic gallium-indium (EGaIn, 75 wt % Ga and 25 wt % In) was purchased from Changsha Kunyong New Materials Co., Ltd. 2-HEA (96%), MTMS (98%), TA (98%), urea (98%), hexadecyltrimethylammonium bromide (CTAB, 99%), 2-hydroxy-4-(2-hydroxyethoxy)-2-methylpropiophenone (2959, 98%), and sodium hydroxide (NaOH, 98%) were obtained from Shanghai Aladdin Co., Ltd. All reagents were used as received without any further purification.

PREPARATION OF LM@TA NANODROPLETS

LM@TA nanodroplets were prepared through an ultrasound-assisted interfacial modification method. Typically, 200 mg of EGaIn and TA were mixed in ethanol (15 mL) at a mass ratio of 10:1. The mixture was sonicated in an ice bath for 1 h using

an ultrasonic cell disruptor (Ningbo Xinzhi) equipped with a 6 mm titanium alloy probe operating at 50% amplitude. This process simultaneously facilitated the breakup of bulk EGaIn into nanoscale droplets and enabled *in situ* surface modification by TA, yielding a uniform dispersion of TA-functionalized LM nanodroplets (LM@TA). The resulting dispersion was centrifuged and washed repeatedly with ethanol to remove unbound TA and collect purified LM@TA nanodroplets. By adjusting the amount of TA in the precursor mixture, LM@TA samples with different surface modification levels were obtained (Table S1).

Fabrication of the PHEA-S/LM@TA hydrogel

The PHEA-S/LM@TA hydrogel was synthesized as follows: 2.21 g HEA and 1.73 g MTMS were dissolved in 3 mL deionized water, where the weak acidity of HEA enabled complete hydrolysis of MTMS. After uniform stirring, 0.76 g urea and 0.19 g CTAB were added, followed by the incorporation of 3.3 wt % LM@TA nanodroplets relative to the total mass of the hydrogel precursor. The mixture was ultrasonicated for 1 min, then 0.0221 g of photoinitiator 2959 was added. The degassed precursor was injected into a mold, UV-irradiated (365 nm, 403 mW cm⁻²) for 4 h, and subsequently thermally polymerized at 80°C for 6 h to obtain the PHEA-S/LM@TA hydrogel. Other hydrogels were prepared by adjusting the mass ratios of LM@TA and the molar ratios of HEA:MTMS (Table S2). A pure PHEA hydrogel, a PHEA-S/LM hydrogel, and a PHEA-S hydrogel without LM@TA were also synthesized under the same conditions for comparison. The PHEA-S/LM@TA hydrogel formulated with 50 wt % solid content, a HEA:MTMS molar ratio of 3:2, and 3.3 wt % LM@TA was used for all studies unless otherwise specified.

Characterizations

The morphology and microstructure of the LM and hydrogels were examined using SEM (Hitachi S-4800 II) and transmission electron microscopy (TEM, Hitachi HT7700). The particle size distribution of LM@TA nanodroplets was measured by DLS on a Malvern Zetasizer system. Chemical structures of the samples were analyzed by FTIR spectroscopy (Nicolet iS10). The surface composition and elemental states of the modified LM and its incorporation into the hydrogel were determined by XPS (Thermo Scientific K-Alpha+). Optical microscopy images were recorded using a Haokang HIF5300 optical microscope. Further details regarding the methods can be found in the supplemental methods.

RESOURCE AVAILABILITY

Lead contact

Requests for further information and resources should be directed to and will be fulfilled by the lead contact, Ximin He (ximinhe@ucla.edu).

Materials availability

This study did not generate new unique reagents. The reagents used here are described in the methods section.

Data and code availability

The data generated during this study are available from the corresponding author upon reasonable request.

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AUTHOR CONTRIBUTIONS

X.S., investigation, data curation, formal analysis, visualization, and writing – original draft; Z.Q., conceptualization, methodology, supervision, project administration, funding acquisition, and writing – review and editing; C.W.Z., formal analysis, visualization, and writing – review and editing; R.K., formal analysis, visualization, and writing – review and editing; Q.X., investigation, methodology, formal analysis, and data curation; N.L., investigation and validation; H.J., investigation and resources; T.J., supervision, resources, and funding acquisition; X.H., formal analysis and writing – review and editing. All authors read and approved the final manuscript.

DECLARATION OF INTERESTS

The authors declare no competing interests.

SUPPLEMENTAL INFORMATION

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