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Review Article

Development of self-healing hydrogels with new functions using nanomaterials

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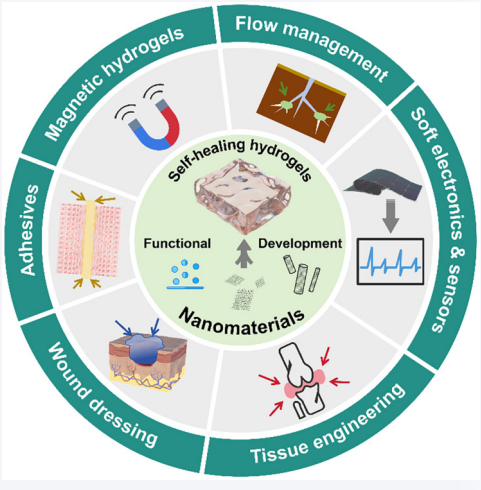
ABSTRACT: Self-healing hydrogels are well-known for their biocompatibility and dynamic networks, and have been considered as new generation smart materials for biomedical applications. However, developing self-healing hydrogels with additional functions through molecular redesign is often complex and costly. Integrating functional nanomaterials with self-healing hydrogels offers a straightforward and efficient strategy to develop new functional self-healing hydrogels. This mini-review focuses on recent advances in self-healing hydrogels functionalized with nanomaterials. It categorizes and discusses these composite materials based on their emerging functions, including wound dressing, tissue engineering, adhesives, magnetic hydrogels, flow management and conductive/sensing systems. The mechanisms of nanomaterial integration, the resulting enhanced properties, and the synergistic performance advantages are discussed. Finally, future perspectives on exploring new functions and potential application scenarios for these nanocomposite hydrogels are presented.



KEYWORDS: self-healing hydrogel, nanomaterials, biomaterials, composite materials

1 Introduction

Self-healing hydrogels are a new type of functional materials with water retention ability, biocompatibility, and shape adaptability [1, 2], and are commonly constructed through intermolecular interaction or dynamic covalent bonds, such as hydrogen bonds [3–5], coordination bonds [6, 7], imine bonds [8–11], borate ester bonds [12–14], and disulfide bonds [15–17]. The resulting dynamic cross-linked networks, which could maintain a balance between destruction and reconstruction, are similar to the extracellular matrix, allowing self-healing hydrogels' application in injectable materials, wound dressing, tissue engineering, and drug delivery [18, 19]. Recently, with the increasing demand for usage and application scenarios, the development of self-healing hydrogels has entered a new stage, which requires the hydrogel to integrate multiple functions [20–22]. However, re-designing and re-synthesizing the molecules toward this target usually increase the difficulty and cost of material preparation and is not favorable for practical applications. Therefore, developing new strategies to enrich and integrate the functions in self-healing hydrogels simply, precisely, and efficiently has become a focus of researchers' attention.

Nanomaterials represent an ideal platform for achieving complex functions, and they have received extensive attention in both scientific research and practical applications [23–25]. At present, nanomaterials could achieve different functions by controlling chemical constitution, morphology, size, and even the spatial sequence, thereby enrich the applications in optics [26, 27], thermodynamics [28, 29] and magnetism [30, 31], catalysis [32, 33], energy storage [34], biomimicry [35, 36], and biological medicine [37, 38]. When combined with the substrate materials, nanomaterials could be dispersed and stabilized as the guest material to enhance the inherent properties or introduce new functions to the host materials. Therefore, incorporating functional nanomaterials into self-healing hydrogels is a straightforward strategy to endow the hydrogels with diverse functions [39–42].

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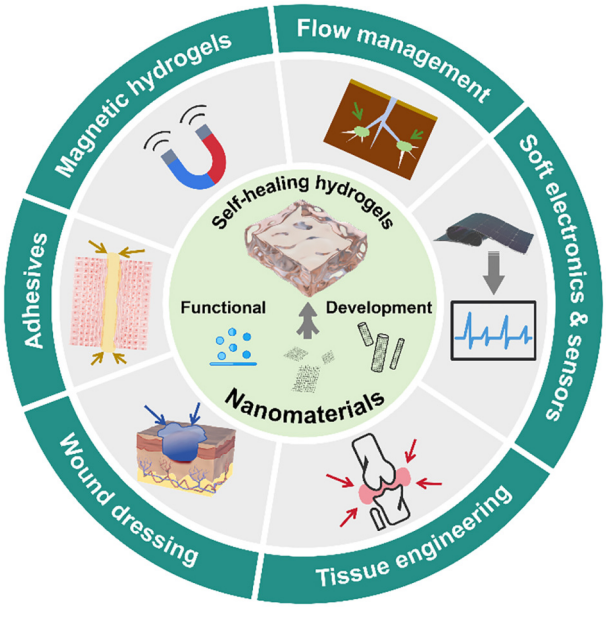
Recently, some studies have combined nanomaterials with hydrogel networks successfully, leveraging the respective advantages of self-healing hydrogels and nanomaterials, and achieving a synergistic effect. The hydrogel-nanomaterial composite materials outperform traditional materials in fields of biomedical materials, tissue engineering and sensors.

In this mini-review, we focus on the development of new functional self-healing hydrogels using nanomaterials, and introduce these composite materials based on the perspective of functional classification. Specifically, some applications that have received widespread attention from researchers will be introduced, including wound dressing materials, implant materials, hydrogel adhesives, magnetic hydrogels, soft electronics and sensing hydrogels, etc. (Scheme 1). We will introduce the mechanism of the combination of nanomaterials and hydrogel networks, discuss the specific functions that nanomaterials bring to hydrogels, and summarize the advantages of hydrogel-nanomaterial composites in terms of performance. Finally, we will identify the existing problems and limitation, discuss the future prospects of using nanomaterials in the development of self-healing hydrogels with new functions, and propose some potential application scenarios.

2 Wound dressing materials

Wound healing is a complex and dynamic process that is influenced by various factors [43]. Internal factors including one's own aging and chronic diseases, and external factors including wound contamination and infection could cause changes in the wound microenvironment, thereby delaying the healing process [44–46]. Traditional wound dressings are unable to respond to changes in the wound microenvironment and cannot adapt to irregular wounds, thus failing to promote the healing process. Recently, self-healing hydrogels have been proven to be safe cell carriers and promote cell migration and proliferation. They can be customized to have specific functions according to the requirements of treating the micro-environment of wound, and the self-healing properties ensure that they can spontaneously adapt to the shape of the wound, thereby promoting healing process [47]. In this new type of wound dressing, the addition of nanomaterials, which play a role as antibacterial and antioxidant materials or carriers for functional drugs, dramatically enhance the effect of wound healing [48].



Scheme 1 Development of self-healing hydrogels with new functions using nanomaterials.

Han and Sun developed a chitosan self-healing hydrogel dispersed with MoS_2 nanosheets [49]. They first conducted ultrasonic mixing on the bulk MoS_2 and the chitosan modified with catechol groups (CSH) in a solution to prepare the MoS_2@CSH nanosheet complexes. The MoS_2 nanosheets dispersed and stabilized in the CSH solution owing to the electrostatic interactions between the sulfur atom and amino group. Meanwhile, the aromatic groups on the modified chitosan have a certain effect in enhancing the hydrophobic interactions. Then, the polyphenolic structures in the CSH could undergo electron transfer under the radiation of near-infrared (NIR) to generate free radicals, thereby forming a cross-linked network (Fig. 1(a)).

The evaluation of the self-healing performance of biomedical hydrogels usually involves both qualitative and quantitative indicators. In terms of macroscopic performance, the self-healing property of hydrogels is manifested as whether they can reassemble into a whole without relying on external intervention after being cut (or broken into pieces by injection and compression). In the quantitative characterization of mechanical properties, the self-healing performance of hydrogels is mainly tested through rheometers. During the rheological experiment, a large strain was applied to break the hydrogel, followed by a small strain to enable it to heal. The circulation process was repeated several times to observe whether the storage modulus (G') and loss modulus (G'') are consistent with those of the original hydrogel and could remain stable.

The MoS_2@CSH hydrogel could heal from fragments into a whole without external force (Fig. 1(b)). After its structure is damaged by high strain shear, it can regain its mechanical properties within a short period of time (Fig. 1(c)). These results indicated that the MoS_2@CSH hydrogel had good self-healing property.

Meanwhile, the unique photothermal conversion effect of the MoS_2 nanosheets caused the increasing of local temperature, which could accelerate the cross-linking reaction process and catalyze the production of hydroxyl radicals (·OH) from H_2O_2. These properties made it possible for the hydrogel to be formed *in situ* at the wound area and achieve broad-spectrum antibacterial effects (Fig. 1(d)). MoS_2@CSH has a very good inhibitory effect on both *E. coli* and *S. aureus* (Figs. 1(e) and 1(f)). Additionally, the active sites of the MoS_2 nanosheets and the polyphenolic structures in the CSH could remove the excessive reactive oxygen species (ROS) to maintain the ROS balance at the wounds. The MoS_2@CSH hydrogel had a better wound healing-promoting effect than the CSH hydrogel without MoS_2 nanosheets, and is significantly superior to the blank group (Fig. 1(g)). The MoS_2 nanosheets facilitated cross-linking reaction and enabled the resulting hydrogel to have antibacterial and antioxidant properties, which demonstrate the rationality of combining nanomaterials with self-healing hydrogels.

Metal-organic frameworks (MOF)-based hydrogels have attracted attention in the fields of sensing and biomedicine [50]. However, the inordinate leakage of metal ions and the insufficient stability of MOF-based materials limit their direct application on wounds. Zhu and Song [51] developed a composite hydrogel using oxidized sodium alginate (OSA), carboxymethyl chitosan (CMCS),

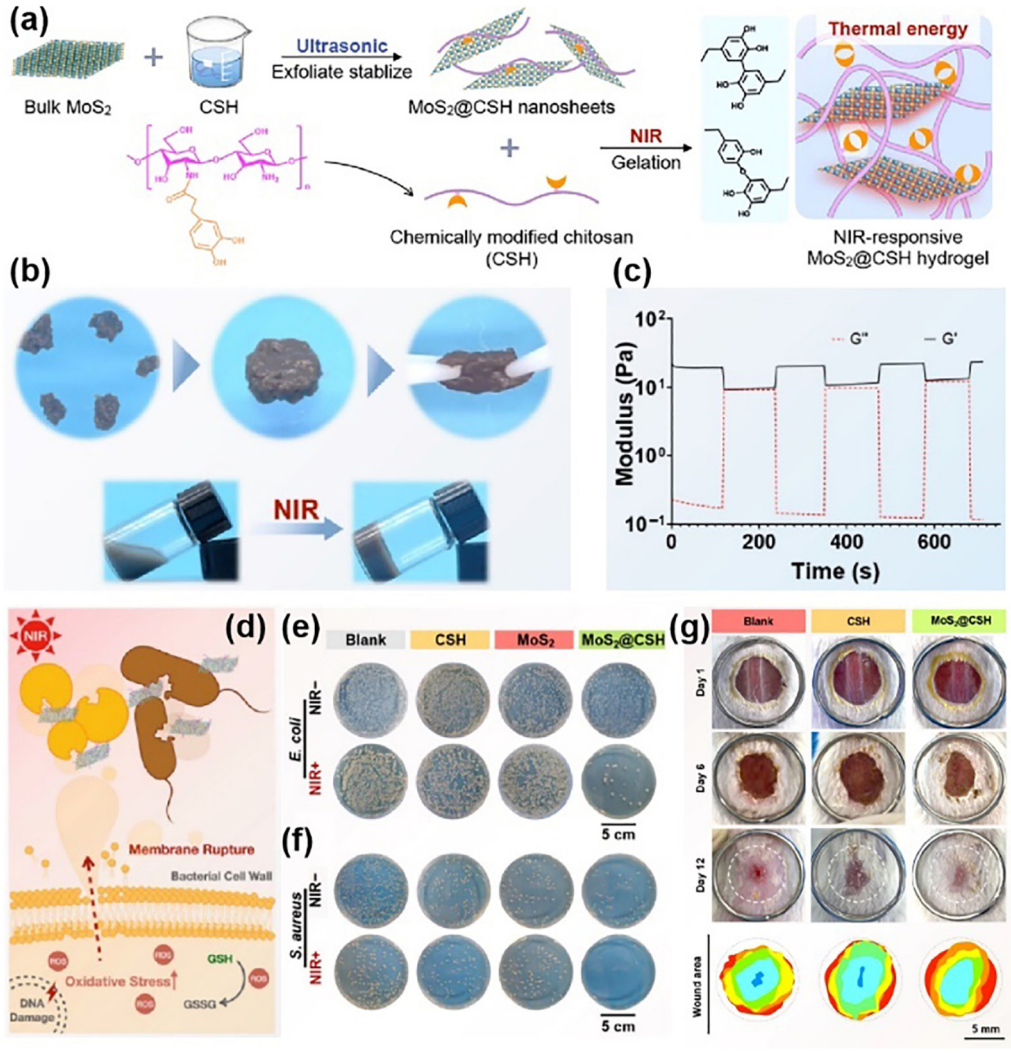


Figure 1 (a) The preparation route of MoS_2@CSH hydrogels. (b) Self-healing properties of MoS_2@CSH hydrogels on different substrates. (c) Rheological properties of the hydrogels dynamic rheological cycle test. (d) Antimicrobial mechanism of MoS_2@CSH. The inhibitory effect on (e) *E. coli.* and (f) *S. aureus.* (g) Representative images of *S. aureus*-infected wounds (blank, CSH and MoS_2@CSH hydrogel on days 1, 6 and 12). Reproduced with permission from Ref. [49], © American Chemical Society 2024.

and nano MOF Cur@Cu-UIO-66-NH_2, which loaded with Cu nanoparticles (NPs) and curcumin to achieve antibacterial and antioxidant functions (Fig. 2(a)).

In the image of transmission electron microscopy (TEM), the Cu-UIO-66-NH_2 (without curcumin) exhibits an octahedral morphology with a size of approximately 200 nm, in which the Cu NPs could uniformly distribute, and the atomic lattice fringes corresponding to the (220) plane of Cu NPs could be observed (Fig. 2(b)). The amino groups on the surface of Cur@Cu-UIO-66-NH_2 could be linked with the OSA through imine bonds, and the hydrogel Cur@Cu-MOF/CMCS-OSA could also be constructed by the dynamic network between OSA and CMCS. In the rheology test, the Cur@Cu-MOF/CMCS-OSA hydrogel, after being damaged by a huge shear strain, was able to quickly restore its mechanical properties under a low shear strain, demonstrating its self-healing properties (Fig. 2(c)). In the antibacterial experiment, the hydrogel loaded with Cur@Cu-MOF significantly inhibited both *S. aureus* and *E. coli* (Figs. 2(d) and 2(e)). In the infected wound model, Cur@Cu-MOF/CMCS-OSA demonstrated a better healing-promoting effect than that of Cu-MOF/CMCS-OSA owing to the synergistic effect of curcumin's anti-inflammatory property and copper nanoparticles' antibacterial property (Fig. 2(f)).

3 Implant and tissue engineering materials

Self-healing hydrogels have emerged as a promising material in tissue engineering due to their unique ability to repair damage and maintain structural integrity [52]. The mechanical properties of self-healing hydrogels are similar to the extracellular matrix, which is conducive to tissue repair. Self-healing hydrogels can be applied onto rough, narrow or deep tissue surfaces due to their inject-ability and self-adaptability [53, 54]. Compared with patch-type materials, they have unique advantages in accelerating cell migration and assisting rehabilitation treatment. As specific functional components, nanomaterials have been added in self-healing hydrogels as active components that assist cell growth in hydrogels or serve as additional physical cross-linking points. The resulting hydrogel-nanomaterial composites could significantly accelerate the tissues' reconstruction process.

Cao and Liao [55] prepared a type of dual-network hydrogel

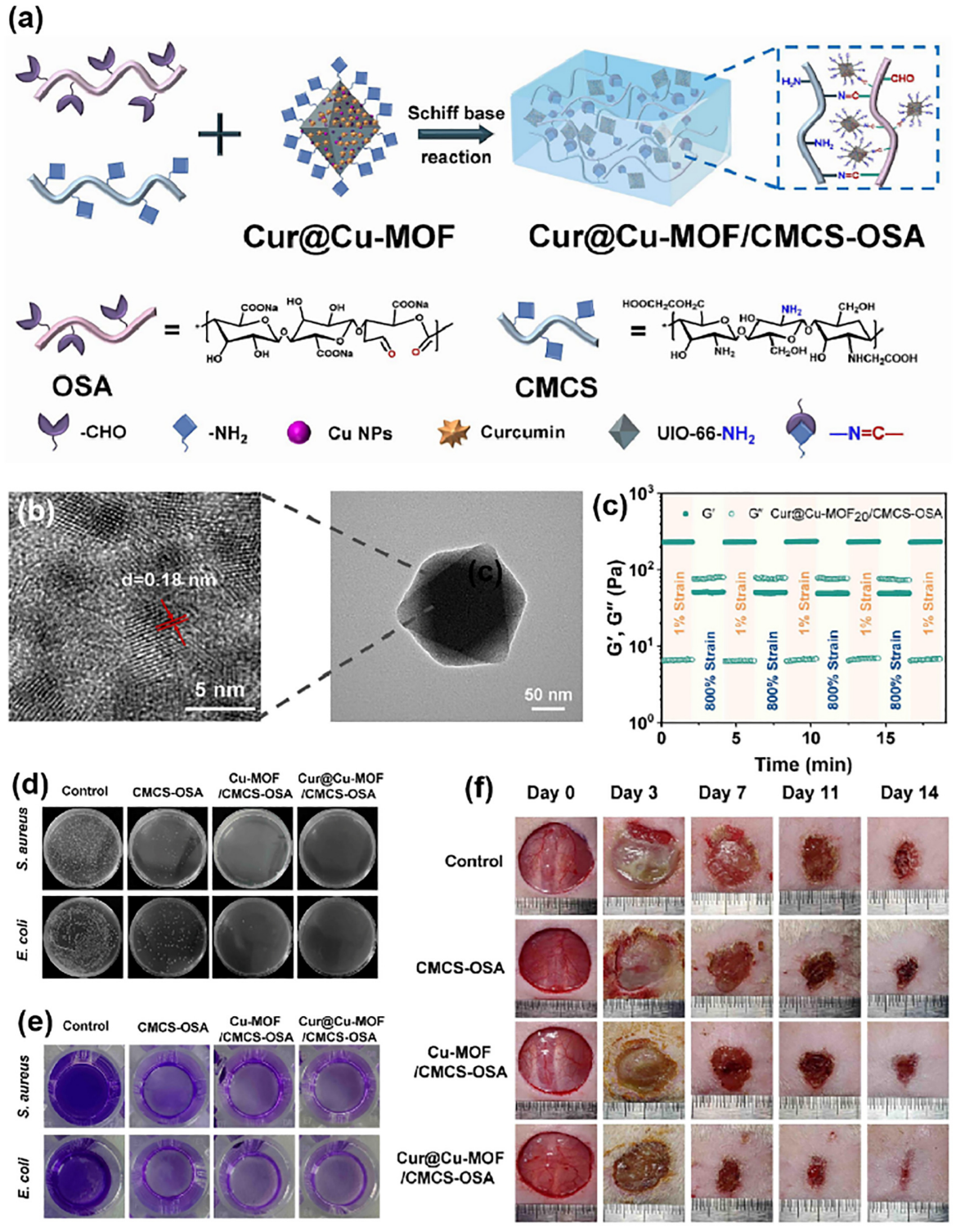


Figure 2 (a) The synthesis process of Cur@Cu-MOF/CMCS-OSA composite hydrogel. (b) TEM images of Cu-UIO-66-NH_2. (c) The recovery situations of the modulus of Cur@Cu-MOF/CMCS-OSA hydrogels after being damaged by a large shear strain. (d) Images of bacterial colonies and (e) crystal violet staining after co-culture of composite hydrogels with *S. aureus* and *E. coli*. (f) Representative images of wound healing within 14 days. Reproduced with permission from Ref. [51], © Elsevier B.V. All rights are reserved, including those for text and data mining, AI training, and similar technologies 2024.

containing hydroxyapatite (HAp). They used oxidized sodium hyaluronate (OHA) and N-carboxyethyl chitosan (CEC) to form the first layer of dynamic network via Schiff's base, and then constructed a covalent cross-linked network containing HAp through the polymerization of polyethylene glycol dimethacrylate (PEGDA) (Fig. 3(a)). The HAp with negative charged surface is physically dispersed by ultrasound and interacts with the positively charged amino groups on the CEC to achieve stabilization, and is then fixed by the dual-network polymer. The dual-network hydrogel loaded with HAp exhibited excellent self-healing

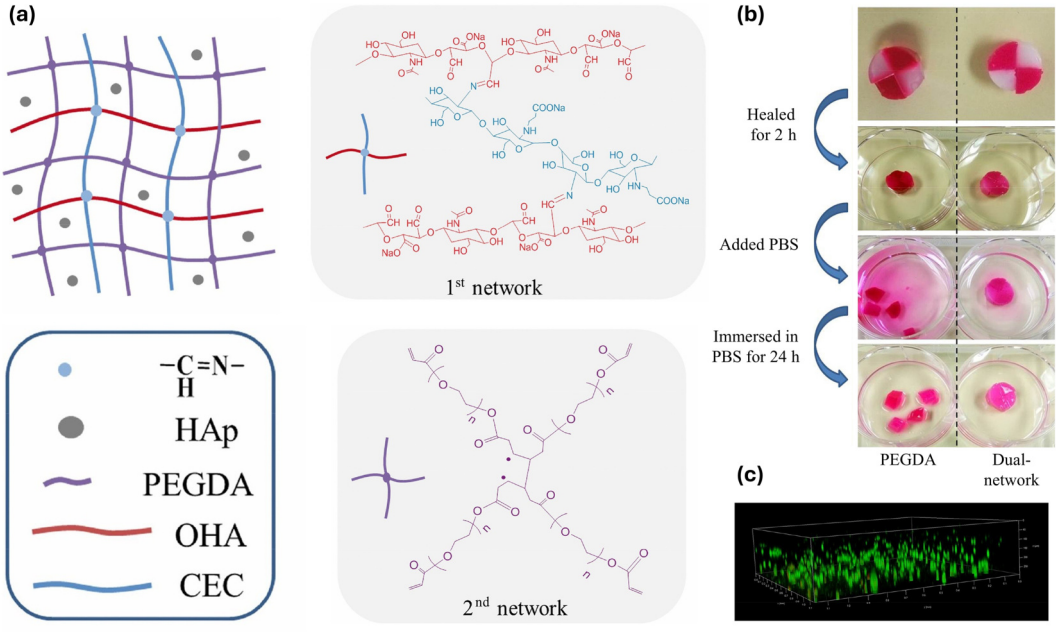


Figure 3 (a) The scheme of dual-network hydrogels. (b) The self-healing properties of dual-network hydrogels. (c) Live/dead staining of the cells in dual-network hydrogels after 3 days. Reproduced with permission from Ref. [55], © Published by Elsevier Ltd on behalf of The editorial office of Journal of Materials Science & Technology 2017.

properties compared with the PEGDA hydrogel (Fig. 3(b)). When used for 3D cell culture, it also demonstrated good biocompatibility (Fig. (c)). These results indicated that this double-layer hydrogel scaffold has broad application potential in the field of bone and cartilage tissue engineering.

Su et al. [56] developed a self-healing hydrogel composed of glycol chitosan (GCS) and oxidized sodium alginate (OSA), which contains calcium phosphate nanoparticles (CaP NPs) for efficient osteoinduction (Fig. 4(a)). The dynamic network was constructed through imine bonds and the mechanical properties, and self-repairing performance of hydrogel could be regulated by adjusting the ratios of amino and aldehyde groups in the two polymers and the amount of calcium phosphate nanoparticles. Compared to the glycol chitosan-oxidized sodium alginate (GCS-OSA) hydrogel without CaP NPs, the composite hydrogel GCS-OSA-CaP can promote the formation of calcium nodules (Figs. 4(b) and 4(c)) and enhance the alkaline phosphatase (ALP) activity of bone marrow mesenchymal stem cells (BMSCs) and the expression of osteogenic-related genes, which facilitates the osteogenic differentiation of BMSCs (Figs. 4(d) and 4(e)). This hydrogel is promising for bone defect repair, drug delivery, and cell encapsulation.

4 Hydrogel adhesives

Polymer adhesives are widely employed for creating temporary or permanent bonds on solid substrates, showing promising potential in applications such as wearable electronics and soft robotics [57–62]. To achieve effective adhesion to common solid surfaces — including glass, metals, and wood — the polymer must contain a sufficient density of functional groups capable of forming interfacial interactions. Moreover, the adhesive itself should exhibit adequate mechanical strength and toughness to withstand external stresses and resist failure or detachment. Although self-healing hydrogels possess the ability to recover from damage to achieve reuse, their practical deployment as adhesives has been limited by the inherent mechanical weaknesses of conventional hydrogel systems. Integrating nanoconfinement strategies into self-healing hydrogel architectures offers a viable pathway to overcome these limitations [63].

The hectorite nanosheets possess the property of forming a nematic liquid crystal structure in water. Zhang, Ikkala and Breu [64] utilized mild shear force to induce the hectorite nanosheets to adopt a more uniform arrangement in the acrylamide monomer solution, and then polymerize through ultraviolet light to prepare nanoconfinement self-healing hydrogel. Due to the nanoconfinement and polymer entanglement, the hydrogel achieved high stiffness (Fig. 5(a)). In the stress-strain test, the strength of the hydrogel significantly increased with the increase in the content of nanosheets (Fig. 5(b)). Due to the re-formation of polymer entanglements at the interface, the hydrogel exhibited a certain self-healing effect, and this self-healing effect was anisotropic. The contact and healing mode in side-by-side arrangement had better performance (Figs. 5(c)–5(e)). This hydrogel had demonstrated excellent bonding performance when interacting with various material substrates (Figs. 5(f)–5(h)).

On the other hands, medical adhesives, commonly known as medical glue, are biocompatible materials used to close wounds and bond tissues [65]. They have become an indispensable tool in modern medicine, partially or completely replacing traditional sutures and staples in many scenarios. The use of medical adhesives could prevent the formation of scars caused by the falling off of surgical sutures and could also reduce the processing time required for wound and tissue suturing. Self-healing hydrogels, as a biocompatible material that can adapt to irregular wounds, have received extensive attention in the development of medical adhesives [66–68]. However, manufacturing self-healing hydrogel adhesives remains a challenging task, as it requires balancing various properties such as scalability, self-healing ability, degradability, biocompatibility and antibacterial performance, which makes the molecular design of polymers quite complicate. As

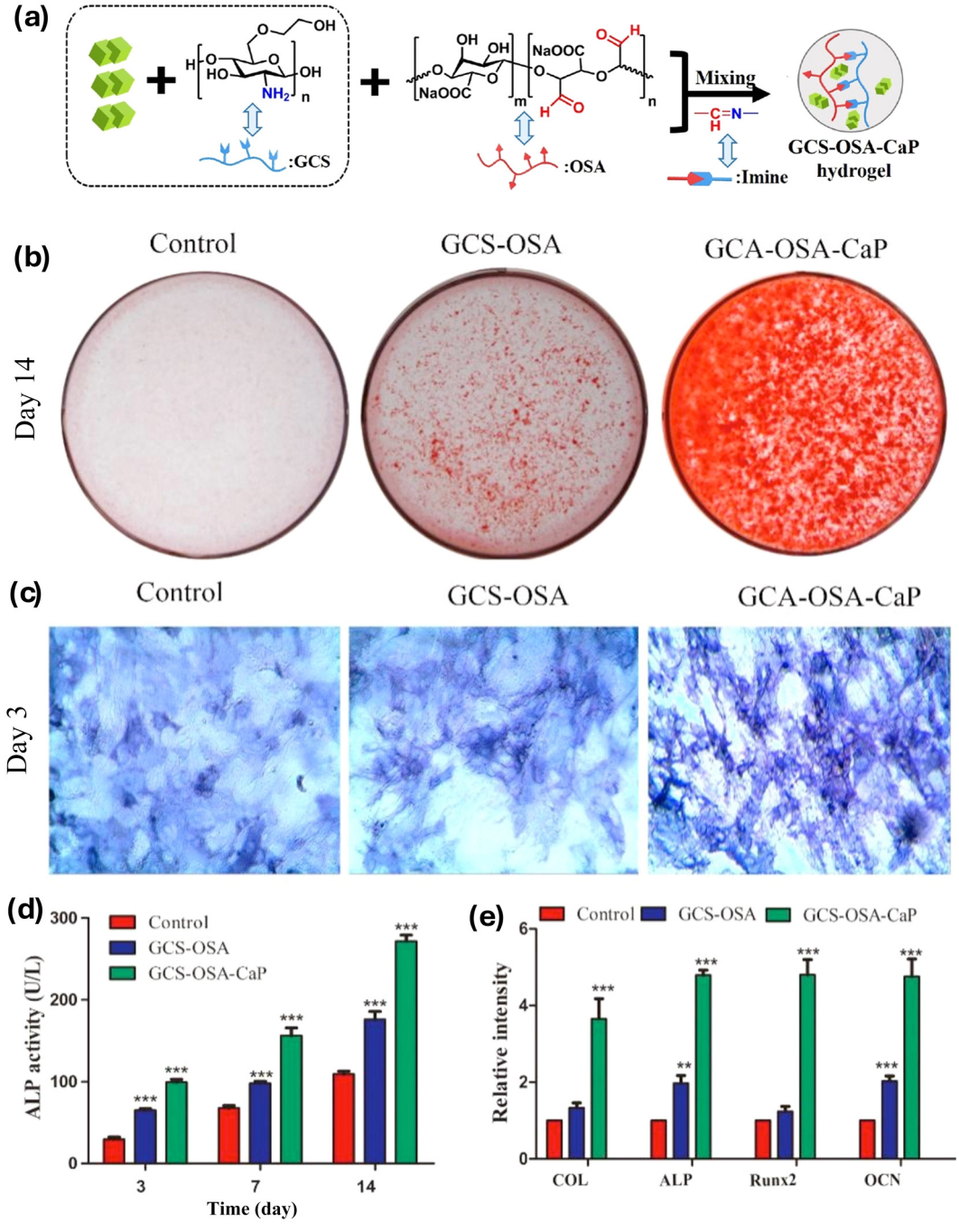


Figure 4 (a) The preparation route of the GSC-OSA-CaP hydrogel. (b) The alizarin staining of calcium deposition by BMSCs in control, GCS-OSA hydrogel and GCA-OSA-CaP group after 14 days of induction. (c) ALP staining after 3 days of induction. (d) Detection of ALP activity in BMSCs. (e) Analysis of mRNA expression of the bone associated markers COL, ALP, RunX2 and osteocalcin (OCN) with qRT-PCR after 21 days of culture. Reproduced with permission from Ref. [56], © Published by Elsevier B.V. on behalf of Chinese Chemical Society and Institute of Materia Medica, Chinese Academy of Medical Sciences 2021.

multifunctional carriers, nanomaterials can provide various solutions for introducing functions into hydrogel medical adhesives, significantly simplifying the preparation of the adhesive materials, and further enhancing their functions.

Shao and Wang [69] developed a type of hydrogel inspired by mussels. Specifically, they modified chitosan (CS) with 3-(3,4-dihydroxyphenyl) propionic acid. Then, they mixed the resulting polymer (DCS) with polyvinyl alcohol (PVA) and subjected the mixture to several freeze-thaw cycles to achieve a self-healing hydrogel (DCS-PVA). The hydrogel was immersed in a $AgNO_3$ solution then reduced by citric acid to produce the hydrogel containing silver nanoparticles and silver clusters (Ag^0@PVA-DCS) (Fig. 6(a)). Scanning electron microscope (SEM) and energy-dispersive X-Ray spectroscopy (EDS) demonstrated that the surface of the hydrogel network had a dense structure of silver nanoparticles or silver clusters, which mainly unevenly deposited but played a role in enhancing the mechanical properties of the hydrogel (Figs. 6(b)–6(d)).

The Ag^0@PVA-DCS was constructed through various reversible interactions such as hydrogen bonds, π-π stacking and cation-π

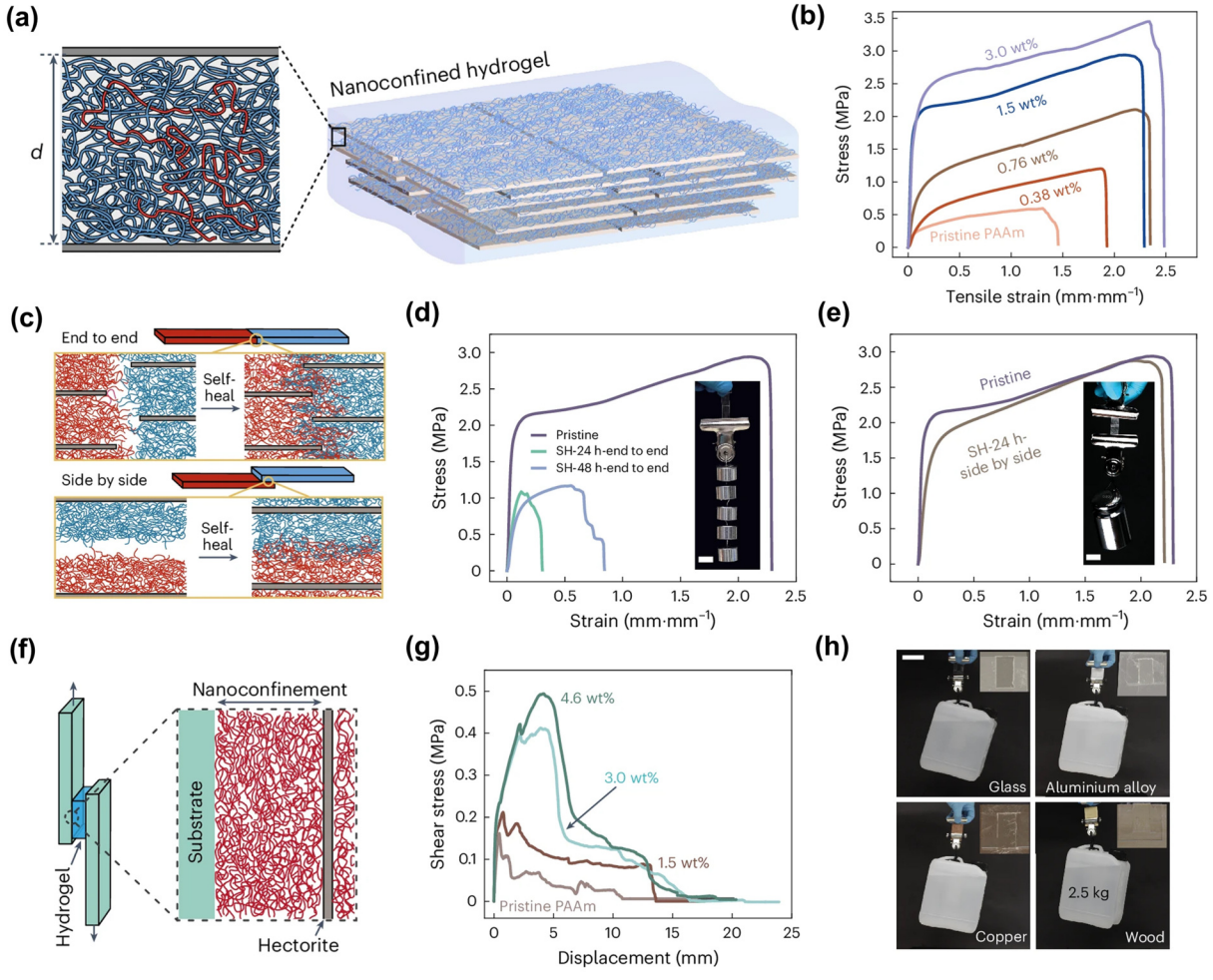


Figure 5 (a) Illustration of the nanoconfinement self-healing hydrogel containing hectorite nanosheets. (b) Tensile stress–strain curves depending on the nanosheet separation defining the confinement. (c) Illustration of self-healing mechanisms based on polymer entanglements along different directions. (d) Tensile curves of nanocomposite hydrogels self-healed (SH) in the *y*–*z* plane (end to end). (e) Tensile curves of nanocomposite hydrogels self-healed in the *x*–*y* plane (side by side). (f) Illustration of lap-shear adhesion test and nanoconfinement at the interface. (g) Nanoconfinement leads to enhanced adhesion. (h) Photographs showing different substrates adhered by hydrogels. Reproduced with permission from Ref. [64], © Liang, C. et al. 2025.

interaction, thus exhibiting excellent self-healing properties (Fig. 6(e)). Meanwhile, the Ag^0@PVA-DCS exhibited excellent antibacterial properties due to the synergistic effect of nano-silver, silver ions and chitosan (Fig. 6(f)). Moreover, Ag^0@PVA-DCS showed good adhesion properties stemming from strong interactions between the surface of objects and its diol moieties (Fig. 6(g)). Therefore, it has potential to be used in the fields of electronic skin and tissue bonding materials. In future, this hydrogel may be upgraded by mixing well-dispersed silver nanoparticles to improve functionality for application.

5 Magnetic self-healing hydrogels

The addition of magnetic nanoparticles to existing materials is a simple way to develop magnetic materials. Magnetic hydrogels have potential in the field of biomedicine such as drug delivery, medical image enhancement, magnetic hyperthermia therapy, and visualization detection [70–72]. However, the hydrogels constructed through stable covalent bonds have poor fluidity and are difficult to be reshaped, which limits their injectability and controllability with magnetic fields. The introduction of self-healing property endows the hydrogel with the injectability and remold ability, allowing it to be driven to a desired location under a magnetic field after loading magnetic nanomaterials, which may enable efficient drug delivery and magnetic heat therapy. The key to achieving magnetic field-driven functionality lies in whether the hydrogels could disperse and combine with the magnetic nanomaterials, preventing the gravitational settling and separation of the internal nanoparticles from the main body of the material.

In 2012, Tao et al. [73] developed a magnetic self-healing hydrogel. This hydrogel is constructed from chitosan and polyethylene glycol capped with benzaldehyde (DF-PEG) via dynamic imine bonds, and combined with the Fe_3O_4 nanoparticles through hydrogen bonds and electrostatic interactions (Fig. 7(a)).

This hydrogel could contact and heal from fragmented parts under the assistance of a magnet (Figs. 7(b)–7(d)). It could pass through a narrow channel with barrier as a whole under the influence of magnetic fields (Figs. 7(e)–7(h)). This study demonstrated the feasibility of loading magnetic nanoparticles onto self-healing hydrogels. Their work conducted early exploration on the magnetic field-driven self-healing hydrogel.

Tao et al. [74] developed a new type of polymer with phosphonic groups and phenylboronic acid groups through the Kabachnick-Fields (KF) reaction and hydrolysis reaction. This functional

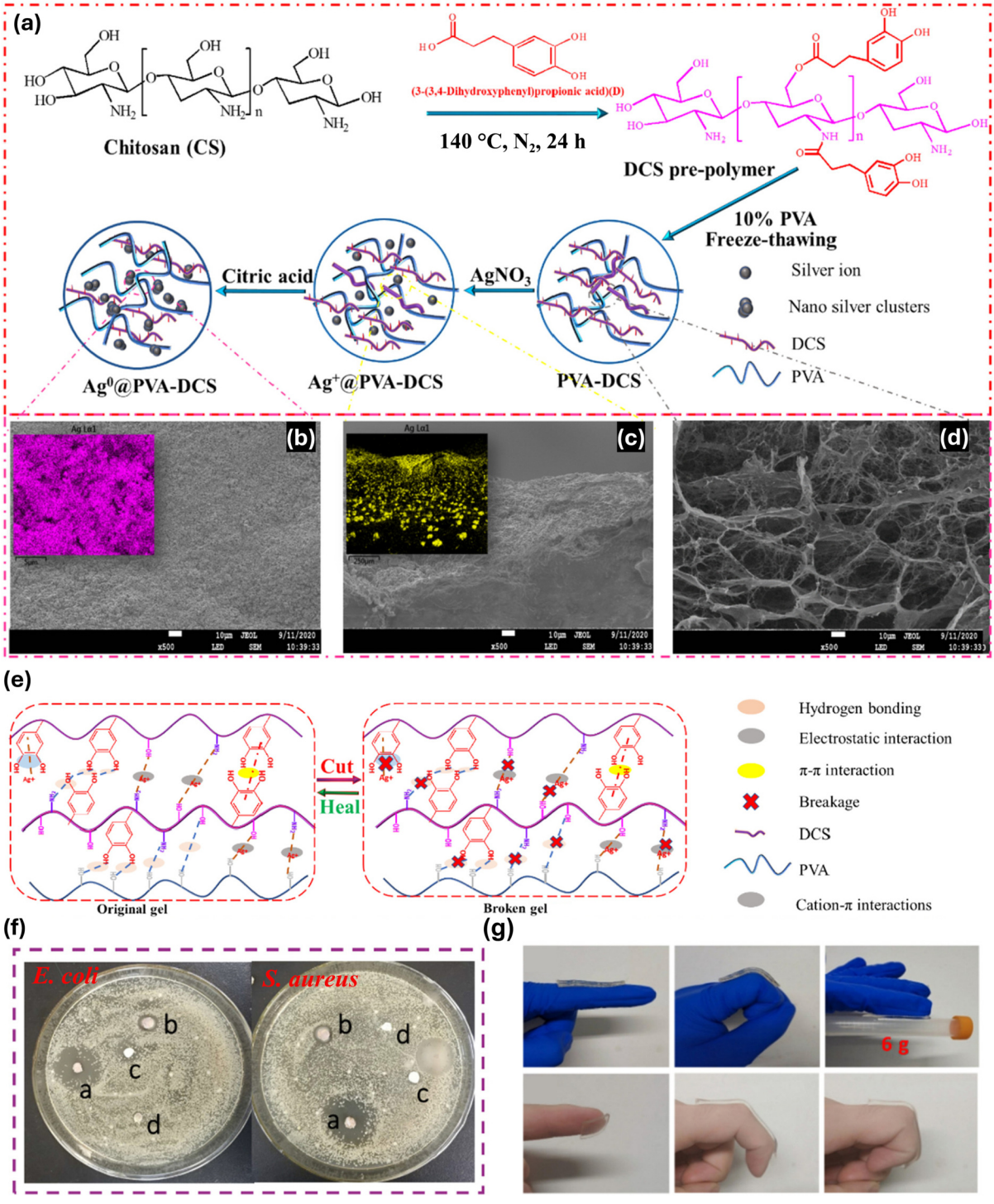


Figure 6 (a) The preparation route of the Ag^0@PVA-DCS hydrogel. (b) SEM and EDS images of the upper surface of Ag^0@PVA-DCS and the distribution of nano-silver clusters; (c) SEM-EDS images of Ag^+@PVA-DCS section; (d) SEM image of the cross-section of the PVA-DCS. (e) Scheme of the healing mechanism of the hydrogel. (f) Antibacterial properties of the samples (a1: Ag^+@PVA-DCS; b1: Ag^0@PVA-DCS; c1: PVA-DCS; d1: neat PVA hydrogel). (g) Adhesion and load bearing of hydrogel on rubber gloves and adhesion of hydrogel to finger joints. Reproduced with permission from Ref. [69], © Published by Elsevier B.V. 2021.

polymer P1 contains phosphonic groups that could combine with the Fe_3O_4 nanoparticles (INOPs), enabling INOPs well dispersed in an aqueous solution. The P1 could also cross-link with PVA in an aqueous solution through dynamic boronic ester bonds (Fig. 8(a)). This self-healing hydrogel could be chopped up into pieces and then healed under the magnetic fields (Fig. 8(b)). Meanwhile, this hydrogel could be heated under the influence of an alternating magnetic field (AMF). By simply changing the content of INOPs, the hydrogel's temperature could be adjusted over a wide range within a shorter period of time (Figs. 8(c) and 8(d)). Furthermore, this INOPs contained hydrogel could influence the nuclear spin-spin relaxation process of protons and reduce the spin relaxation time (T_2) during magnetic resonance imaging (MRI) (Figs. 8(e) and 8(f)), which could improve the quality of MRI. These results

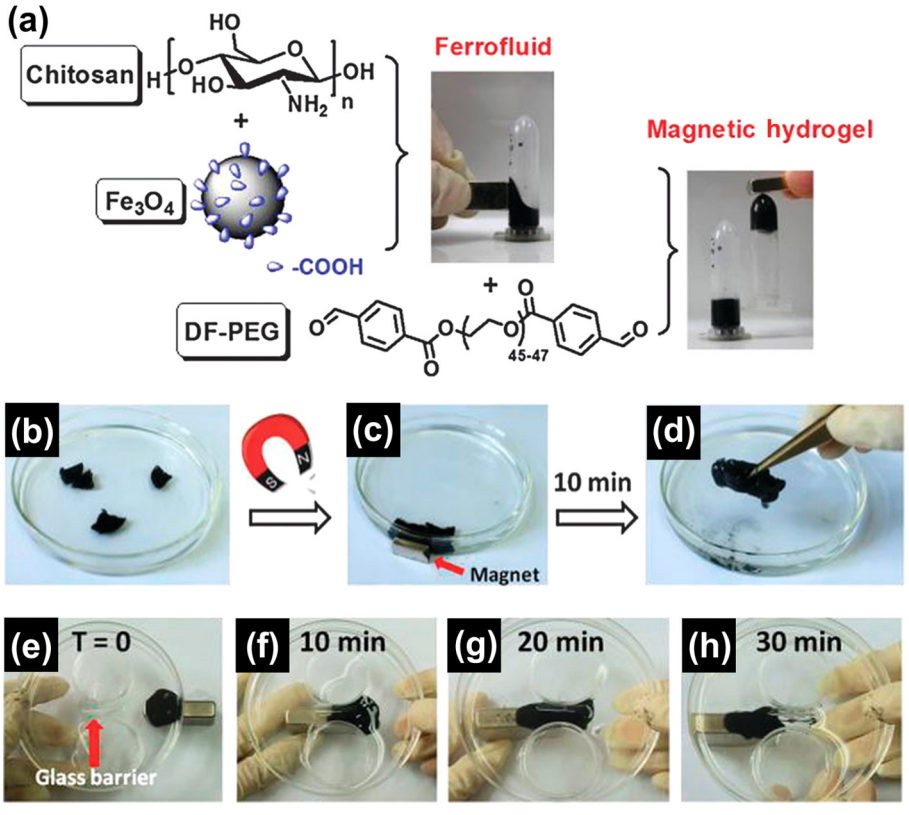


Figure 7 (a) The preparation route of magnetic hydrogel. (b)–(d) The healing effect of the magnetic hydrogel under the influence of magnetic fields. (e)–(h) The magnetic hydrogel passed through narrow channels with barrier as a whole. Reproduced with permission from Ref. [73], © The Royal Society of Chemistry 2012.

indicated that this hydrogel has the potential to be used as an injectable MRI contrast agent.

Li et al. [75] developed a self-healing hydrogel based on oxidized pectin and chitosan. After being oxidized by sodium periodate, the aldehyde groups generated on the pectin could form dynamic imine bonds with the amino groups on chitosan (Figs. 9(a) and 9(b)).

The hydrogel could be attracted by a magnet after loading the γ-Fe_2O_3 nanoparticles, which were dispersed through mechanical stirring (Fig. 9(c)). The obtained hydrogel-γ-Fe_2O_3 exhibited excellent self-repairing and injectable properties (Fig. 9(d)). After being loaded with the anti-cancer drug 5-fluorouracil (5-FU), the hydrogel-γ-Fe_2O_3 could continuously release the drug for up to 12 h. This hydrogel exhibited excellent properties such as injection capability, self-repairing ability, magnetism, high biocompatibility, and anti-cancer characteristics, making it a promising candidate carrier for anti-cancer drugs.

6 Soft electronics and sensors

Hydrogels have attracted significant attention in the research of wearable or implantable flexible electronic devices as a possible material substrate due to their mechanical properties being similar to those of various biological tissues [76–78]. Self-healing hydrogels typically possess high water content, tissue adhesiveness, biocompatibility, and self-repairing capabilities. They overcome the drawbacks of traditional hydrogels, such as performance degradation after structural rupture, and insufficient extensibility and shape adaptability. However, due to the lack of the conductive structures or a relatively small amounts of conductive ions, self-healing hydrogels, like traditional hydrogels, have the inherent problem of low ionic conductivity, which limits their application in actuating devices and high-sensitivity sensing. To address this issue, nano-scale conductive materials (such as metallic nanowires or nanoparticles, carbon-based nanomaterials, MXene-based nanomaterials, and some nano-scale intrinsic conductive polymers) have been incorporated into hydrogels [20, 79–81]. The composite materials combining conductive nanomaterials and self-healing hydrogels exhibit excellent electrical conductivity and tissue-like mechanical flexibility, and have broad application prospects in the field of soft electronics and sensors.

Metal nanoparticles are common conductive nanomaterials. The outermost electrons of metal atoms are highly delocalized and they can move directionally within the metal under the influence of an external electric field, ensuring that the metal itself has a relatively high electrical conductivity. However, in self-healing hydrogels, due to the dynamic properties of the polymer networks, the metal nanoparticles will precipitate under the influence of gravity, altering their dispersion properties and preventing the formation of tunneling electric conduction between the nanoparticles and significantly changing the overall electrical conductivity of the hydrogel. Therefore, achieving the uniform dispersion of nanoparticles in the self-healing hydrogel is crucial.

Son and Shin [82] developed an injectable tissue-interfacing prostheses composed of conductive hydrogel (IT-IC hydrogel), which is constructed with irreversible biphenyl bond, reversible hydrogen bonds, metal-π and metal-carboxylate weak interaction (Fig. 10(a)). In this study, the disruption and reformation of non-covalent interactions and the rearrangement of biphenyl covalent bonds enable the hydrogel to dissipate the shear stress during

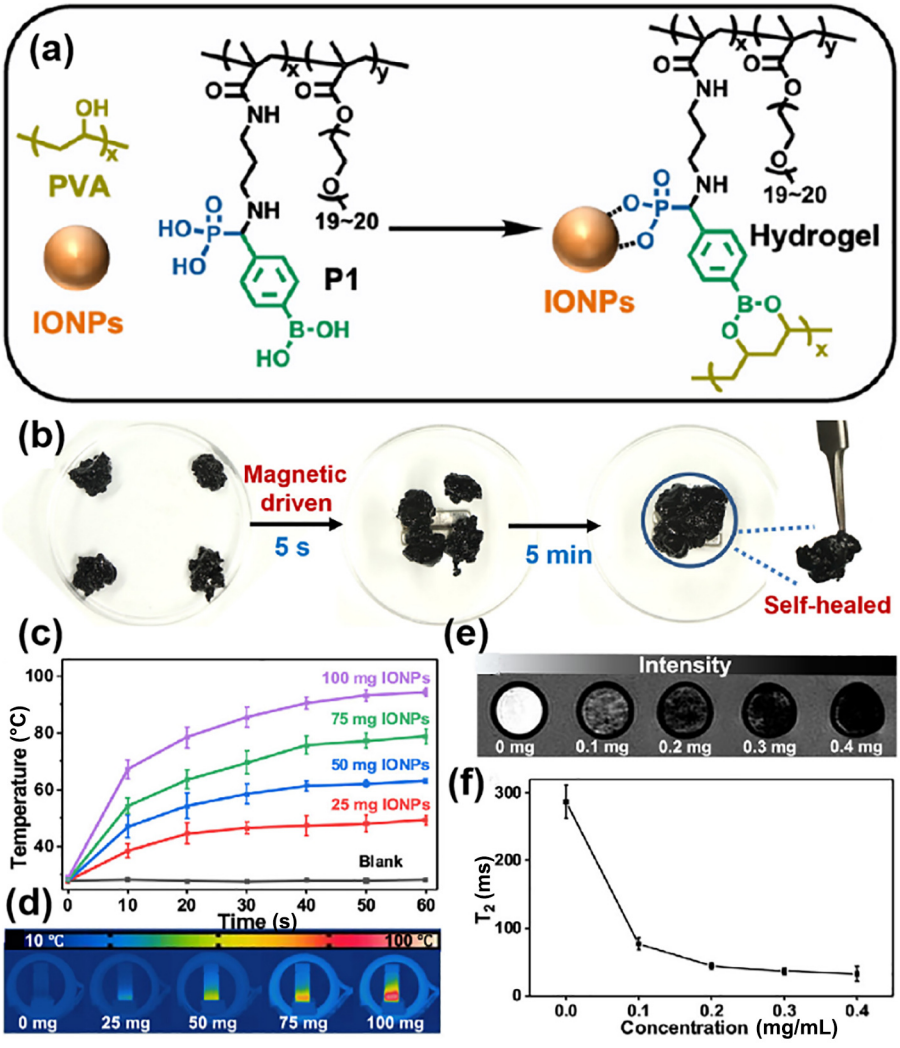


Figure 8 (a) The preparation route of the hydrogel and the mechanism for dispersing IONPs. (b) Magnetic-driven healing of hydrogels. (c) Temperature changes and (d) infrared images of hydrogels with different contents of IONPs under alternating magnetic fields. (e) MRI images and (f) T_2 values of hydrogels containing different amounts of IONPs. Reproduced with permission from Ref. [74], © American Chemical Society 2021.

injection and maintain its mechanical properties while using. The Au nanoparticles (AuNPs) are generated *in situ* by the reduction of Au^{3+} within the cross-linked network. These AuNps have a strong coordination with the phenylboronic acid groups in the polymer. This interaction stabilizes the AuNPs, enabling the IT-IC hydrogel to possess continuous conductive properties (Fig. 10(b)). This injectable conductive implant material can fill the rough surface of injured muscle tissue and assist in the transmission of electrical signals within the muscle tissue, thereby accelerating the regeneration of the tissue and the restoration of its functions. With the help of the closed-loop robot-assisted rehabilitation (C-RAR) system comprising IT-IC hydrogels, a robot, a treadmill and self-healing stretchable cuff and epimysial electrodes, rats with volumetric muscle loss (VML) injury surgery were able to resume normal walking under neural stimulation (Fig. 10(c)). This work developed the IT-IC hydrogel that established a two-way interaction between muscles and the surrounding nerve tissues, providing a new material for the rehabilitation treatment of patients with chronic tissue injuries.

Carbon-based nanomaterials have conductive properties similar to those of metals because the carbon atoms form large planar structures in the form of Sp2 hybridization, allowing the free electrons to delocalize on the plane [83, 84]. However, carbon-based nanomaterials have extremely strong hydrophobicity. They are difficult to be dispersed in the hydrogel system and are prone to re-aggregation, thereby altering the electrical conductivity of the material. Therefore, efficiently dispersing carbon-based nanomaterials in hydrogels is the key to developing carbon nanomaterial-hydrogel composite materials.

Li and Yao [85] developed a conductive hydrogel loaded with carbon nanotubes (CNTs). The CNTs are dispersed in the gelatin solution through hydrophobic interactions. Then, the sulfobetaine methacrylate (SBMA) monomer is added to this dispersed system for polymerization, and a dynamic polymer network is constructed through electrostatic interactions (Fig. 11(a)). Due to the electrostatic properties of the poly(sulfobetaine methacrylate) (PSBMA) network not being swollen in water and having excellent self-healing properties (Fig. 11(b)), the introduction of CNTs endowed the hydrogel with relatively sensitive changes in electrical signals, enabling it to distinguish different stresses and strains (Figs. 11(c) and 11(d)). Meanwhile, for the movements of different parts of human body (such as the bending of fingers and wrists),

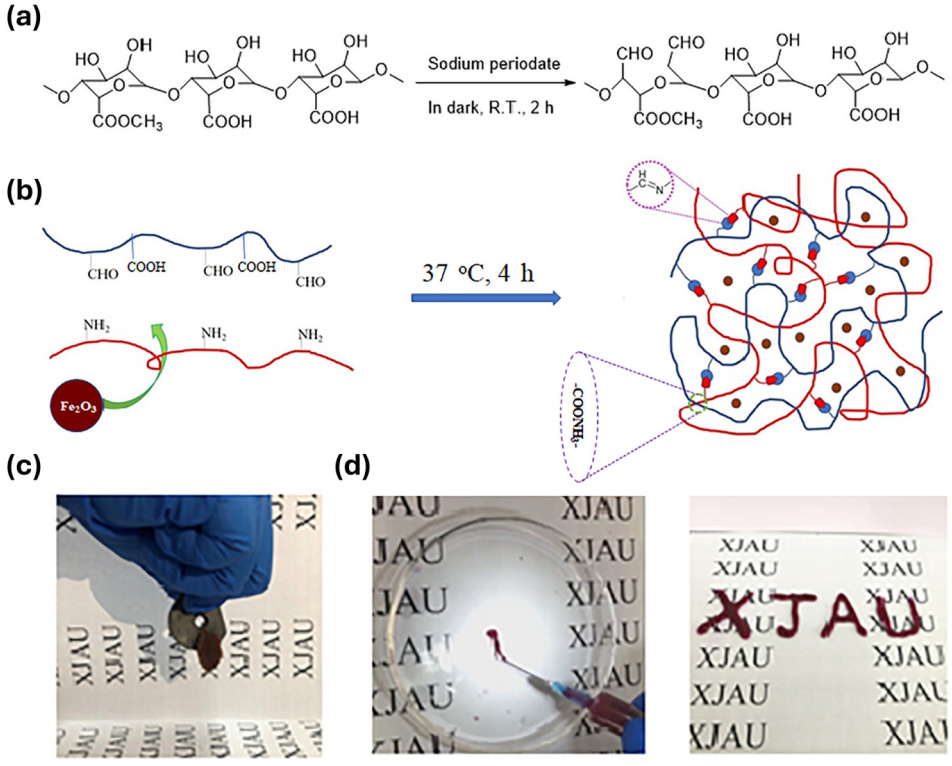


Figure 9 (a) The synthetic route of oxidized pectin. (b) The preparation route of the γ-Fe_2O_3 nanoparticles contained hydrogel. (c) The attraction of magnets to the hydrogel. (d) The hydrogel can be injected through a needle and form specific letters due to its self-healing property. Reproduced with permission from Ref. [75], © Elsevier B.V. All rights reserved 2020.

this hydrogel sensor can generate different electrical signals that can be clearly distinguished and recognized, and it is effective in both air and water (Figs. 11(e)–11(h)).

Conductive polymers, such as polyacetylene [86, 87], polyaniline [88, 89], polypyrrole [90, 91], polythiophene [92] and PEDOT/PSS (poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate)) [93–95], have been broadly used for the development of flexible electronic devices. These polymers possess conjugated π-bond frameworks, which provide conditions for the delocalization and movement of electrons. Conductive polymers could introduce a large number of charge carriers (electrons or holes) through further doping, enabling their electrical conductivity to reach the level of metals. Adding nanoscale conductive polymers to self-healing hydrogels not only enhances their electrical conductivity but also may form more cross-linking sites, thereby improving their mechanical properties.

Wang and Dong [96] developed a muscle-inspired self-healing hydrogel. This hydrogel is formed through the coordination interaction between polyacrylic acid (PAA) and Fe^{3+}, as well as the electrostatic interaction between PAA and sharp-featured polyaniline nanofibers (PANI NFS), and the electrostatic interaction between PAA and Fe^{3+}, to create a dynamically cross-linked network (Fig. 12(a)). The PANI NFs were dispersed by sonication in an aqueous solution of acrylic acid (AA) followed by polymerization of AA to produce PAA-PANI binary networked-hydrogel (PPBN hydrogel) (Fig. 12(b)). This hydrogel exhibits excellent extensibility and can maintain certain mechanical properties even after multiple damages and self-healing processes (Figs. 12(c) and 12(d)). PANI NFs are a type of conductive material. The surfaces of their nanoscale structures exhibit jump conduction and tunnel conduction phenomena. Therefore, this hydrogel not only can sense strain (Fig. 12(e)) but also has a sensitive temperature sensing ability (Fig. 12(f)). The sensors made from this hydrogel can identify a wide range of temperature changes, and the experimental results can be repeated even when the sensors are repeatedly exposed to cold and hot conditions (Fig. 12(g)).

7 Self-healing hydrogels for other applications

With the in-depth research on self-healing hydrogels, some functions that were previously not considered in traditional fields are now being discovered. Injecting cold water and gas into the ground to obtain geothermal energy is a method for improving energy utilization efficiency [97]. However, prolonged fluid injection could lead to the formation of fractures and high-permeability channels within the formation, resulting in fluid loss and insufficient recovery. By injecting the hydrogel underground, the sealing of cracks and high-permeability channels could be achieved, which is an effective method to improve the flow control effect of the geothermal system [98]. The self-healing hydrogels constructed by reversible dynamic bonds possess the ability of self-repair. They can repair the fractures caused by shear stress and overcome the drawbacks of traditional hydrogels that use toxic crosslinking agents. Adding nanomaterials to hydrogels used for fluid management can form additional physical crosslinking points, significantly enhancing their mechanical strength, thereby maintaining the integrity of the gel under mechanical force and prolonging its service life.

Guo et al. [99] developed a polyacrylamide hydrogel. This hydrogel was cross-linked through dynamic disulfide bonds and exhibited the properties of shape reformation and self-healing (Fig. 13(a)). The images obtained from the SEM show that the silica

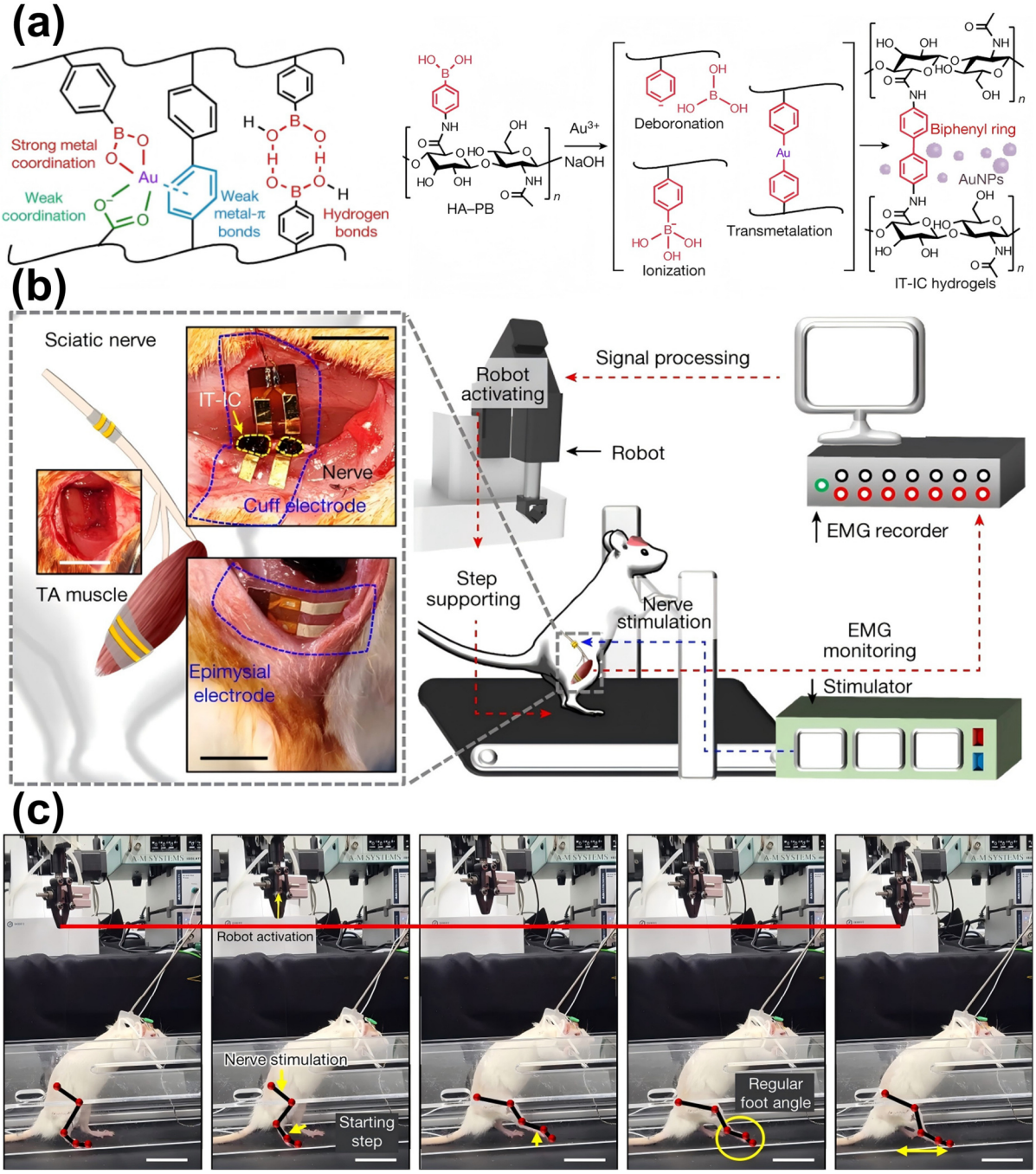


Figure 10 (a) The scheme of the structure, interaction and preparation route of the hydrogel. (b) The scheme and image of the C-RAR system with IT-IC hydrogels, a robot, a treadmill and self-healing stretchable cuff and epimysial electrodes. (c) Rats with VML injury surgery were able to resume normal walking with the assistance of the C-RAR system. Reproduced with permission from Ref. [82], © Jin, S. B. et al. under exclusive licence to Springer Nature Limited 2023.

nanoparticles are attached to the cross-linked polymer network, providing a supporting framework and thereby enhancing the overall strength of the gel system (Fig. 13(b)).

The compressive modulus of the hydrogel in the compression test and the storage modulus in the rheological test increased significantly (Figs. 13(c) and 13(d)). In addition, the silica nanoparticles in the hydrogel not only increase the cross-linking sites, but also enhance water retention. The viscosity, storage modulus and loss modulus of the system remain basically unchanged at 140 °C. After 100 days of aging, the gel system does not dehydrate, and the cross-linking structure remains stable under high-temperature conditions (Fig. 13(e)). The injection pressure test showed that the *in-situ* formed hydrogel has excellent adhesion to the pipeline. Under high-pressure water flow, it can only break through the structure of the hydrogel and will not push the entire hydrogel out, which is conducive to achieving a strong water-blocking effect (Fig. 13(f)).

8 Summary and outlook

Self-healing hydrogels, as a new type of dynamic material, are attracting increasing attention in the research field due to their

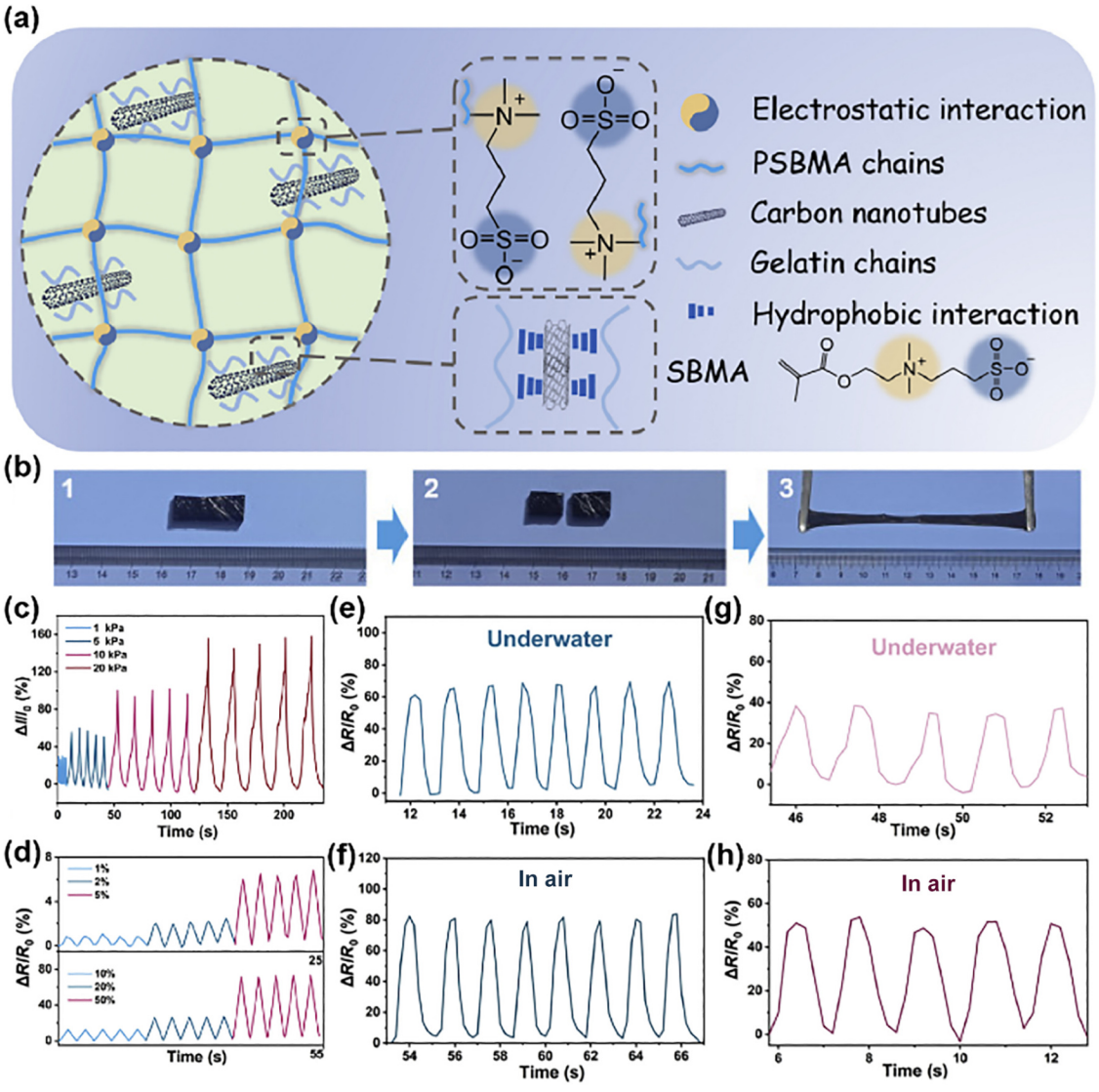


Figure 11 (a) The scheme of conductive hydrogel loaded with CNTs. (b) The self-healing properties of the hydrogel. The changes in electrical signals of the hydrogel under different (c) stresses and (d) strains. Hydrogels as strain sensors to monitor finger bending (e) underwater and (f) in air. Hydrogels as strain sensors to monitor wrist bending (g) underwater and (h) in air. Reproduced with permission from Ref. [85], © Elsevier Ltd. All rights reserved 2021.

similar mechanical properties to biological tissues, as well as their good biocompatibility and shape adaptability. In this mini review, we introduce the use of nanomaterials in the development of functional self-healing hydrogels, and classify these composite hydrogels according to different functions. As definable functional additives, nanomaterials have enriched the application scenarios (such as wound dressings, tissue engineering materials, magnetic hydrogels, flow control materials, flexible electronic devices and sensors) of self-healing hydrogels in a relatively simple way, avoiding the complex molecular design and related cost-increase, and providing new strategies for the development of new functional materials.

However, as a carrier material, self-healing hydrogels are not perfect. For instance, when used in flexible electronic devices or sensors, changes in the moisture content within the hydrogels can lead to issues such as variations in conductivity, making these materials difficult to meet the requirements for long-term stable use. Therefore, researchers should consider methods for maintaining the performance of self-healing hydrogels over the long term, or explore application scenarios suitable for existing materials. Some packaging methods, as well as underwater sensors or in-body implant sensors, are directions that can be explored. In addition, during the use of hydrogels, the issue of leakage of nanomaterials should also be taken into consideration, especially in the biological field. This might lead to serious health risks. By modifying the nanomaterials with functional groups that can interact with the hydrogel network, it might be possible to enhance the stability of the nanomaterials and reduce their leakage. Moreover, although nanomaterials have powerful functions and potential for large-scale production, the biological safety and environmental safety of nanomaterials added to wound dressings, tissue engineering materials, or medical adhesives have not been verified, or have been proven to be harmful. When selecting nanomaterials to develop hydrogel-nanomaterial composites, researchers should take the safety of the materials into consideration, and also verify the biological safety and environmental safety of the composite hydrogels.

Overall, integrating these two emerging materials, nanomaterials

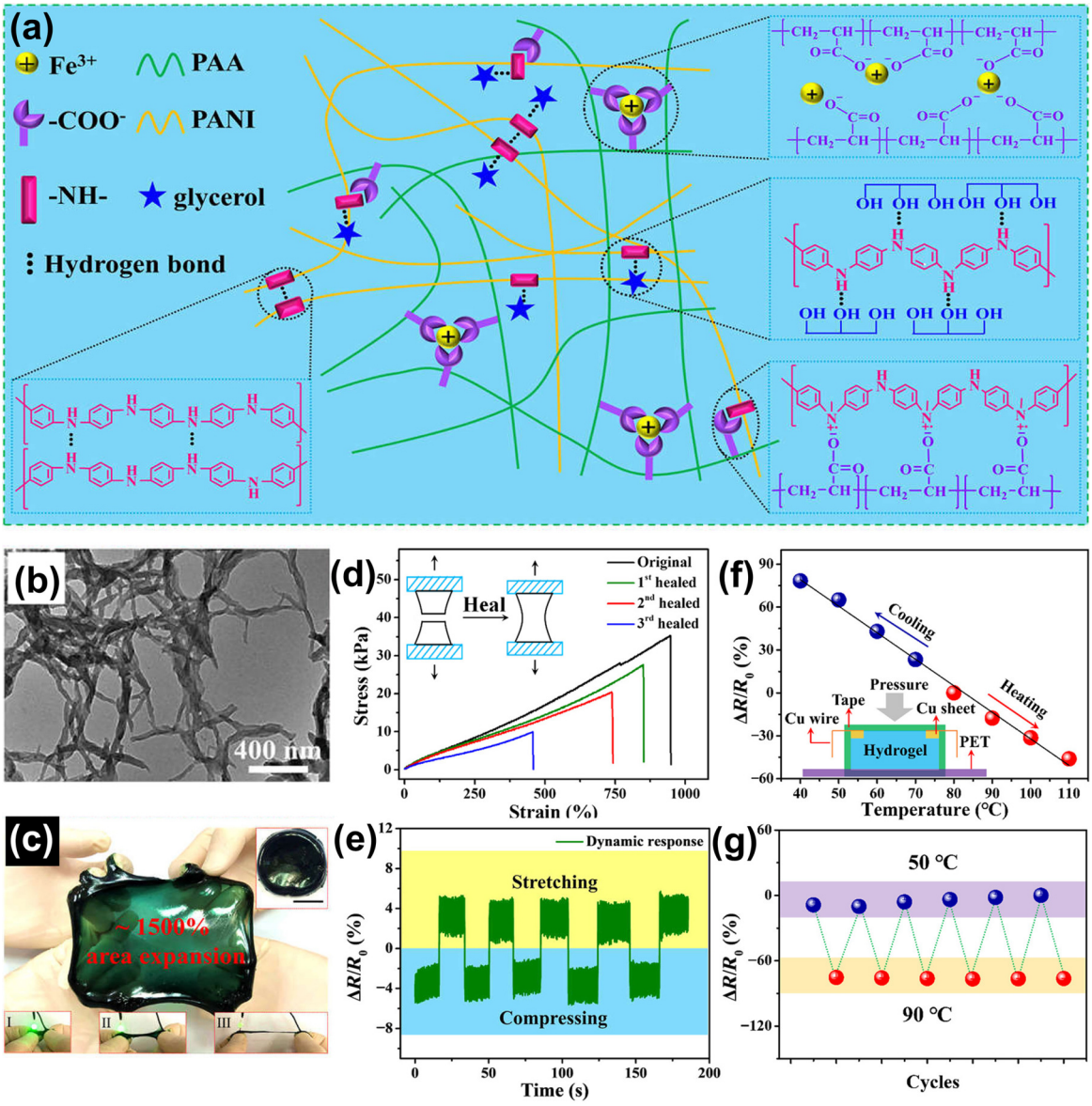


Figure 12 (a) Schematic structure of the multifunctional hydrogel and its reversible interactions. (b) TEM image of PANI NFs. (c) Illustration of the biaxial area expansion (1500%) and uniaxial stretch ability of the hydrogel. The upper right inset shows the original circular sample (scale bar: 2 cm). Three bottom insets show luminance variation of a green light-emitting diode (LED) during stretching, where states I, II, and III refer to 0%, 300%, and 600% strain, respectively. (d) Mechanical performance of PPBN-hydrogel after three successive breakage-healing processes. (e) Dynamic response of the sensor to tiny tensile strain (5%) and compressive strain (5%). (f) Normalized relative resistance variation ($\Delta R/R_0$ (%)) of the sensor as a function of temperature (from 40 to 110 °C). (g) Reproducible temperature-discrimination capacity of the sensor during repetitive cold (50 °C) and heat (90 °C) source approaching. Reproduced with permission from Ref. [96], © American Chemical Society 2019.

and self-healing hydrogels, represents a new approach for developing new functional materials. Nanomaterials have brought many possibilities for expanding the functions and improving the performance of hydrogels. At present, the research on applying nanomaterials to the development of functional self-healing hydrogels is still in its infancy. In the future, we may witness technological breakthroughs of nanomaterials@self-healing hydrogels in various fields, and even attempts and applications in some unexpected areas.

Data availability

Not applicable.

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Declaration of competing interest

All the contributing authors report no conflict of interests in this work.

Author contribution statement

Z. Y. L. drafted the manuscript. G. Z. L. and L. T. reviewed and revised the manuscript.

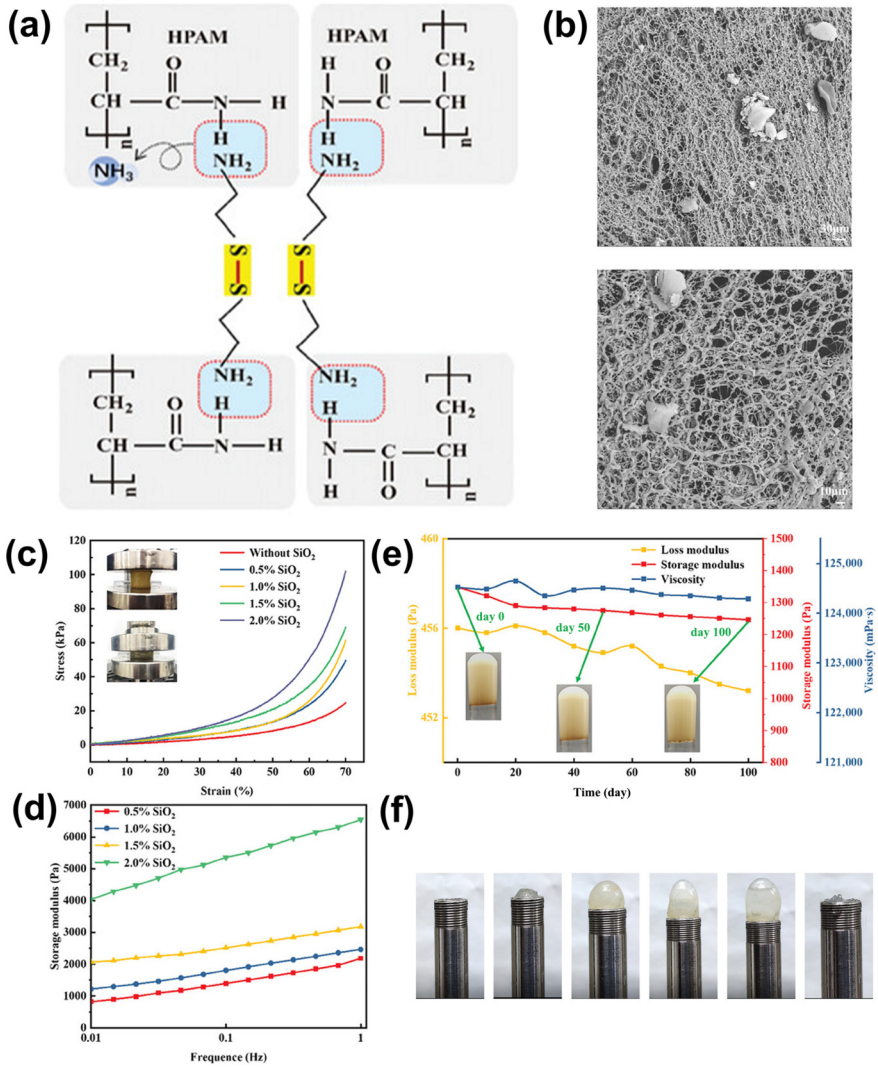

Figure 13 (a) The scheme of polyacrylamide hydrogel cross-linked by disulfide bonds. (b) Microstructure of the cross-linked gel system. The changes in the (c) compressive modulus and (d) storage modulus of hydrogels with different contents of SiO_2. (e) Change of gel properties over time in an environment of 140 °C. (f) Breakthrough pressure experiment process. Reproduced with permission from Ref. [99], © Wiley-VCH GmbH 2024.

Informed consent

Not applicable.

Ethics statement

Not applicable.

Use of AI statement

None.

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