Review



# Recent Advances of Conductive Hydrogels for Flexible Electronics

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Keywords: Conductive hydrogel; Mechanical strength; Tissue engineering; Flexible electronics

*Review*

# Recent Advances of Conductive Hydrogels for Flexible Electronics

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**Abstract:** Conductive hydrogels combine the properties of both hydrogels and electrical conductivity, making them soft, flexible, and biocompatible. These properties enable them to conform to irregular surfaces, stretch and bend without losing their electrical conductivity, and interface with biological systems. Conductive hydrogels can be utilized as conductive traces, electrodes, or as a matrix for flexible electronics. Exciting applications in sensors, tissue engineering and human-machine interaction have been demonstrated worldwide. This review comprehensively covers the progress in this field, focusing on several main aspects: functional materials, performance improvement strategies, and wearable applications in human-related areas. Furthermore, the major approaches and challenges for improving their mechanical properties, conductivity, and long-term stability are systematically summarized.

**Keywords:** conductive hydrogel; mechanical strength; tissue engineering; flexible electronics

## 1. Introduction

Hydrogels are three-dimensional crosslinked networks of hydrophilic polymers capable of holding large amount of water within their structures. They can absorb and retain water while maintaining their structural integrity, with water contents typically exceeding 90 %. The structure and properties of hydrogels can be tailored for specific applications by adjusting parameters such as polymer composition, crosslinking density, and network architecture. This tunability allows hydrogels to exhibit a wide range of characteristics, including varying degrees of swelling, mechanical strength, and biocompatibility, making them valuable materials as excellent substrates, carriers, or scaffolds in certain fields. Conductive hydrogels endow the hydrogel with electrical conductivity while retaining its hydrophilic and swellable nature [1–4]. These properties enable them to conform to irregular surfaces, stretch and bend without losing their electrical conductivity, and interface with biological systems, making them applicable in tissue engineering, biosensors, bioelectronics, soft robotics, and wearable electronics.

Comparing to traditional inorganic conductive materials, conductive hydrogels achieve complementary characteristics of conductive materials and hydrogels. Taking advantage of their outstanding conductivity, conductive hydrogels have emerged as ideal candidates for flexible electronic skin sensors [5,6]. Additionally, conductive hydrogels possess outstanding biological properties such as self-healing, biocompatibility, adhesion, and antibacterial properties, which present unique advantages when interfacing with biological tissues or organisms for signal tracking [7,8]. Currently, conductive hydrogels have been widely demonstrated in the fields of wearable sensors, biomedical devices, actuators, soft electronics, touch panels, energy storage devices, sustained drug release systems, to controlled stimulus-responsive drug delivery systems. Conductive hydrogels are also utilized in human applications such as wound dressings [9,10] and human tissue

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engineering [10]. In these fields, conductive hydrogels act as bridges, enhancing communication and electrical coupling between normal and damaged tissues [11–14].

Though conductive hydrogels have gained significant attention due to their unique combination of properties, they also come with certain disadvantages, including limited mechanical strength, limited durability, potential toxicity, and compatibility issues. The purpose of this paper is to review the progress of conductive hydrogels and their applications as expanding devices and functional materials (Figure 1). In the first section, we briefly introduce the fabrication of conductive hydrogels. Based on the types of conductive principles, we classify conductive hydrogels into three categories: electronic conductive hydrogels, nanoparticle conductive hydrogels, and ion conductive hydrogels. Next, we describe the key properties of conductive hydrogels and their enhancement strategies. In the third section, we focus on some important applications of conductive hydrogels in tissue engineering and flexible electronic devices. Finally, we summarize the issues and limitations of conductive hydrogels and provide prospects for their future development.

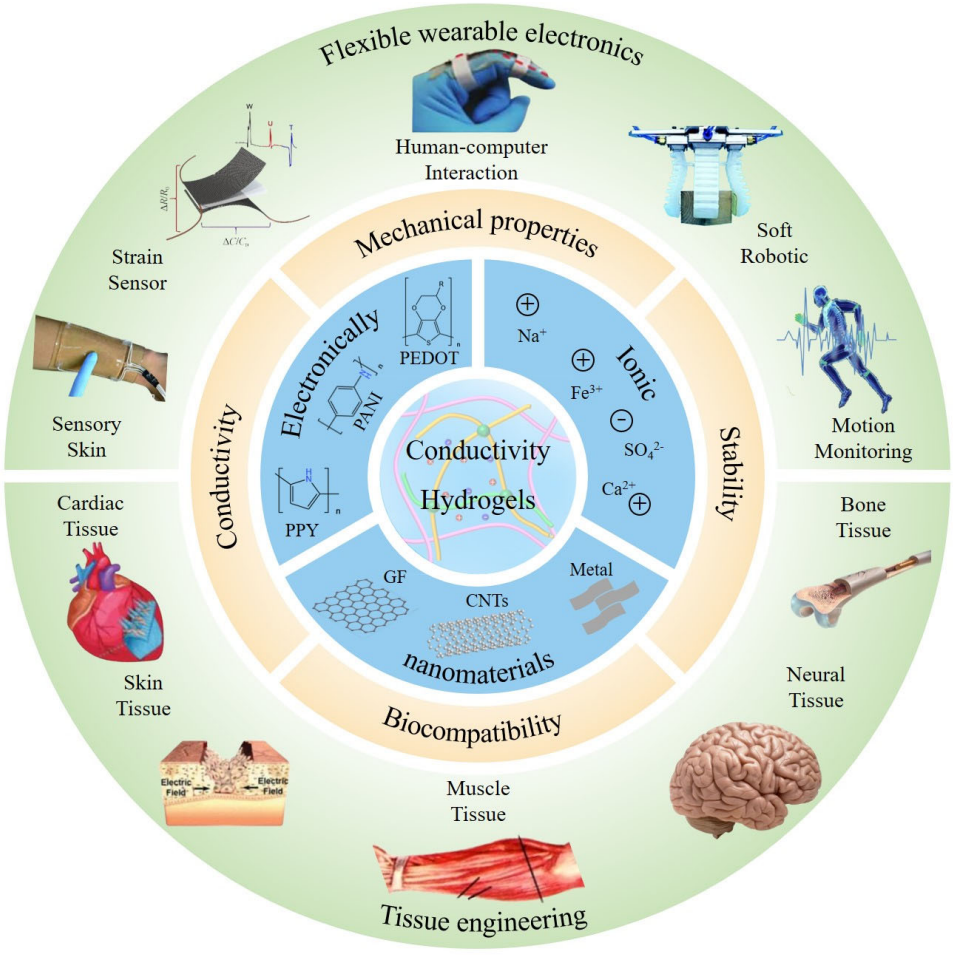


**Figure 1.** Classification, properties and applications of conductive hydrogels. The various applications of the conductive hydrogels include: Sensory Skin [15], Strain sensor [16], Human-computer interaction [17], Soft robotic [18], Motion monitoring [19] for Flexible wearable electronics, and Cardiac tissue [20], Skin tissue [21], Muscle tissue (from the Internet), Neural tissue (from the Internet), Bone tissue (from the Internet) for Tissue engineering.

## 2. Fabrication of conductive hydrogels

Conductive hydrogels have tremendous potential in the fields of flexible wearable technology and tissue engineering. Many research efforts have been made to design and fabricate various conductive hydrogels. According to conductive fillers, conductive hydrogels can be classified into three main categories (Figure 2): electronic conductive hydrogels, nanoparticle conductive hydrogels,

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and ion conductive hydrogels. The conductivity mechanisms, advantages, disadvantages, and research progress of each type of conductive hydrogel are introduced carefully.

*2.1 Electronic conductive hydrogels*

Conductive polymers (CPs) typically consist of carbon atoms and p-conjugated electron systems, endowing them with the ability to conduct electrons. Compared to other conductive materials, they offer advantages such as tunable electronic conductivity, flexibility, and biocompatibility. The conjugated structure comprises local σ bonds and π bonds. During polymerization, overlap of p orbitals between π bonds induces electron redistribution [22]. Conductive polymers, characterized by their unique p-conjugated structures, find application in electron transfer, including polyaniline (PANI), polypyrene (PPy), poly (3,4-ethylene dioxythiophene), and polythiophene. They are commonly utilized as fillers in polymer frameworks or conductive hydrogels shown in Figure 2a [23,24]. As shown in Figure 2b, conductive polymer-based hydrogels primarily rely on electron transfer as charge carriers for conduction, thus exhibiting good environmental stability and conductivity [25]. In recent years, conductive polymer-based hydrogels have garnered significant attention due to their stable chemical properties and excellent conductivity. A microwave and pH-responsive conductive hydrogel was developed using in-situ polymerization to incorporate polyaniline within a poly(N-isopropylacrylamide) (PNIPAM) gel matrix [26]. This innovative approach aimed to create a hydrogel that responds to both microwave radiation and changes in pH. Initially synthesized in 1988, poly (3,4-ethylene dioxythiophene) (PEDOT) faced limitations due to its poor water solubility. However, its conductivity was successfully enhanced through doping with carbon-based materials or protonic acids. A composite of polystyrene sulfonate (PSS) and PEDOT, known as PEDOT: PSS, has since found wide-ranging applications in various fields [27]. Hydrogel sensors capable of accurately detecting minor movements such as pulses were fabricated by incorporating PEDOT: PSS into a hydrogel matrix [28]. Zhang et al. explored the synthesis of a novel conductive γ-GM-P hydrogel composed of γ-poly (glutamic acid) (PGA) and PEDOT: PSS [28], adjusting the PEDOT: PSS content to modulate pore size, electromechanical properties, and swelling rate. Improvements in both capacitance and mechanical performance were achieved by bridging polyaniline with PEDOT using phytic acid [29]. Embedding polyaniline particles in the three-dimensional (3D) network of paper-like PEDOT facilitated rapid electron transfer, resulting in high capacitance performance. Efforts to enhance hydrogel conductivity involved introducing proton-doped polyaniline into the hydrogel matrix, significantly improving its electrical properties. Challenges persisted with polypyrene due to its hydrophobic nature when combined with hydrogels. Addressing these challenges, researchers introduced a novel freeze-polymerization method, resulting in excellent conductive materials with high conductivity, mechanical strength, and super-elasticity [30]. This method involved vertically freezing a mixed solution of polyvinyl alcohol (PVA), aniline (ANI), and an initiator, inducing PVA to form a three-dimensional ordered honeycomb structure growing along the direction of ice crystals. Moreover, researchers focused on synthesizing conductive composite hydrogels tailored for flexible wearable applications. One approach [31] involved the polymerization of polyaniline (PANI) around aromatic polyamide nanofibers-polyvinyl alcohol (ANF-PVA) templates, shown in Figure 2c, resulting hydrogel exhibited outstanding mechanical properties, strain sensitivity, stability, and durability.

*2.2. Nanoparticle conductive hydrogels*

Comparative to conductive polymers, the utilization of conductive composite hydrogels (CPS) incorporating nanoparticles as the conductive medium facilitates simplified fabrication procedures. Integration of nanoparticles into hydrogels, encompassing both metal and carbon-based variants boasting elevated conductivity levels, engenders the formation of a permeable particle network. This network effectively augments both the electrical and mechanical attributes inherent to hydrogels.

Metal nanoparticles represent the preferred choice for fashioning functional conductive hydrogels, lauded for their notable conductivity, optical prowess, catalytic proficiency, and ease of manipulation. Diverse materials, including metal and metal oxide nanoparticles, nanowires, and

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nanorods, have garnered substantial adoption in recent years. Noteworthy examples include the integration of gold and silver nanoparticles, alongside silver nanowires and liquid metal variants. Traditional metal nanoparticles are often fraught with challenges pertaining to aggregation and sedimentation within hydrogel matrices. To circumvent such issues, Lin et al. [26,32] devised a methodology involving the immobilization of silver nanoparticles (Ag) onto tannic acid (TA)-modified cellulose nanocrystals (CNCs). This approach capitalizes on the outstanding dispersibility and compatibility exhibited by CNCs with hydrophilic polymers, thus facilitating the uniform dispersion of Ag within the polyvinyl alcohol (PVA) matrix. Consequently, this technique yielded boronic acid-silver/tannic acid cellulose (PB-Ag/TA@CNCs) hydrogels distinguished by their commendable electrical and mechanical characteristics. Furthermore, Lee et al. [33] successfully incorporated silver nanowires (AgNWs) into polyacrylamide hydrogels, thereby yielding highly pliable hydrogel micro-patterned electrodes. The inclusion of AgNWs markedly enhanced the electrical conductivity of the hydrogel microelectrodes, achieving resistances as low as 109 Ω. While the integration of metal nanoparticles fortifies the electrical and mechanical aspects of hydrogels, it may impart certain drawbacks, including potential impacts on their electrical properties and heightened production costs, thus posing constraints on large-scale applicability. Carbon-based nanoparticles offer multifaceted utility, serving as both active materials for energy storage and conduits for energy transfer networks. This diverse category encompasses graphene oxide (GO) (Figure 2d), carbon nanotubes (CNTs) (Figure 2e), carbon fibers, metal particles (Figure 2f), and others, all of which exhibit commendable electrical conductivity, environmental robustness, and biocompatibility. Notably, graphene and carbon nanotubes have emerged as pivotal elements in constructing three-dimensional conductive networks atop polymer substrates. Leveraging their conjugated structures, these materials afford efficient pathways for electron transfer while simultaneously boasting commendable mechanical properties attributable to their expansive surface functionalities and high specific surface areas. As a result, they have found wide-ranging utility in biosensing applications and the development of flexible wearable bioelectronics. In contrast to their metal counterparts, carbon-based nanomaterials demonstrate remarkable stability in wet environments, thereby fostering their integration within conductive nanocomposite hydrogels. Nevertheless, the innate hydrophobic nature of carbon-based materials predisposes them to agglomeration and poor dispersion in aqueous environments, thus precluding the formation of uniform and stable conductive networks within hydrogel matrices. To address these inherent challenges, various strategies have been proposed, such as functionalizing carbon-based materials or incorporating hydrophilic substrates to enhance their compatibility with hydrogels. A chemically-physical hybrid hydrogel incorporating carboxyl-functionalized multi-walled carbon nanotubes (MWCNTs) was reported [34], achieving exceptional and sustained electrical conductivity through electrostatic interactions. Similarly, reduced graphene oxide (rGO) synthesized via polydopamine (PDA) was utilized to ensure homogeneous dispersion within a polyacrylamide (PAM) hydrogel matrix, resulting in commendable mechanical and electrical attributes [35]. Other strategies, including the integration of graphite-derived graphene oxide (GO) and reduced graphene oxide (rGO) [36], as well as the introduction of materials such as cellulose nanofibers (CNF), hydrophilic polymers, and compounds [37], have demonstrated efficacy in addressing these challenges. Noteworthy is the engineering of a multifunctional conductive hydrogel comprising a polyvinyl alcohol-borax matrix supplemented with CNF and carbon nanotubes (CNTs), referred to as a polyvinyl alcohol-borax hydrogel/carbon nanotube-cellulose nanofiber (CNT-CNF) [38]. This composite material exhibited outstanding electrical conductivity, reaching levels as high as 0.1 S/cm due to the incorporation of CNTs. The synergy between carbon-based nanomaterials and hydrogel matrices in composite materials offers a blend of electrical and physical attributes. However, challenges remain due to the limited dispersibility and elevated costs associated with nanomaterials such as CNTs and GO, which may impede large-scale production efforts.

As elucidated above, the non-biodegradability of carbon nanomaterials within biological contexts poses a significant impediment to their utility in tissue engineering applications [39,40]. Consequently, burgeoning interest has shifted towards two-dimensional nanomaterials boasting

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superior performance metrics, such as BP nanosheets or MXene nanosheets. BP nanosheets, for instance, exhibit conductivity levels akin to graphene-like semiconductors (100 S/cm−1) and can be metabolized into benign phosphorus elements within physiological environments. For instance, synthesized PDA-modified BP nanosheets as conductive additives for GelMA hydrogels, effectively modulating the neural differentiation of mesenchymal stem cells (MSCs) by Xu et al. [41]. Similarly, two-dimensional transition metal carbides/nitrides (MXene) have garnered substantial interest since their inception in 2011. Characterized by exceptional hydrophilicity, conductivity, and tunability, MXene has found widespread utility in the domain of conductive hydrogels. Tao et al. [42] detailed the synthesis of a nano-composite hydrogel predicated on a novel peroxide-modified MXene nanosheet (p-Ti3C2Tx). The resultant hydrogel showcased uniform distribution of MXene throughout its structure, thereby facilitating comprehensive coverage of electron channels across the gel network. Consequently, MXene nanosheets, typifying two-dimensional transition metal carbide nanomaterials, have found application within electroconductive hydrogels matrices for 3D cell culture and bioprinting endeavors [43], courtesy of their commendable conductivity, biodegradability, and biocompatibility. In essence, while metal nanoparticles bolster the electrical characteristics of conductive hydrogels, challenges pertaining to aggregation persist. Carbon-based nanoparticles, on the other hand, exhibit immense promise albeit with hurdles related to dispersion. The advent of biodegradable two-dimensional nanomaterials presents a promising avenue for surmounting concerns associated with non-biodegradable carbon nanomaterials, particularly within the realm of tissue engineering

### *2.3. Ion conductive hydrogels*

Unlike electronically conductive hydrogels, ionically conductive hydrogels closely mimic ion transport in biological tissues and cells, making them better suited for wearable sensors and simulating human soft tissues. The mechanism behind ionic conduction involves the electrical conductivity of ions through free movement. This can be broadly classified into three categories: metal salts (such as FeCl3/FeNO3, NaCl, KCl, LiCl, AlCl3, TbCl3, CaCO3), acids (such as H3PO4, HCl, H2SO4), and ionic liquids (such as 1-ethyl-3-methylimidazolium acetate [37]. The mainstream approach involves dissolving conductive ion salts (such as NaCl, LiCl, etc.) into hydrogels, thereby imparting them with high ionic conductivity, flexibility, and stretchability. Due to the absence of additional conductive polymers and nanoparticles, conductivity is entirely dependent on ionic conductivity, making them safer and highly transparent. This unique advantage has positioned them as a popular choice in the field of flexible bioelectronics, becoming a hotspot in recent years for the preparation of conductive hydrogels. A study explored the synthesis of a self-healing, stretchable conductive hydrogel based on cellulose nanocrystals (CNC) - Fe3+ and polyvinyl alcohol (PVA)/polyvinylpyrrolidone (PVP) [44], achieving a maximum tensile strain of 1,160% and demonstrating excellent self-healing properties. Additionally, another type of conductive hydrogel was developed, where a β-cyclodextrin (β-CD)/polyvinyl alcohol (PVA) hydrogel (sn-hydrogel) was immersed in a polyacrylic acid (PAA)/potassium chloride solution to prepare dn-hydrogel [45]. In alkaline or neutral media, the ionization of carboxyl groups on PAA chains gradually increased, facilitating the migration of K+ and Cl-, leading to a sharp decrease in resistance.

In addition to enhancing ionic conductivity, the incorporation of salt ions can serve other purposes, such as improving the mechanical properties and frost resistance of hydrogels. As shown in Figure 2g, improvements in the mechanical properties of hydrogels were achieved by introducing Ca2+ to regulate interchain interactions, forming metal chelation bonds [19,46]. Conductive hydrogels based on electrolytes have found extensive applications, particularly in supercapacitors. Moreover, salt ions not only contribute to conductivity but also enhance hydrogel performance in extreme environments. A freeze-resistant, mechanically robust PVA/glycerol/sodium alginate/CaCl2 double network ion hydrogel was successfully prepared, combining various properties to broaden the scope of conductive hydrogel applications across diverse scenarios [47]. Similarly, $Fe^{3+}$ also are utilized to enhance the bonding (Figure 2h). However, high concentrations of ions in wearable bioelectronic devices, while beneficial for sensor conductivity, may pose risks to cells and tissues. To

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address this concern, highly conductive salt solutions were encapsulated within polyethylene glycol matrices to fabricate conductive hydrogels with both high conductivity and biocompatibility, suitable for bio-matched electronic interfaces [48].

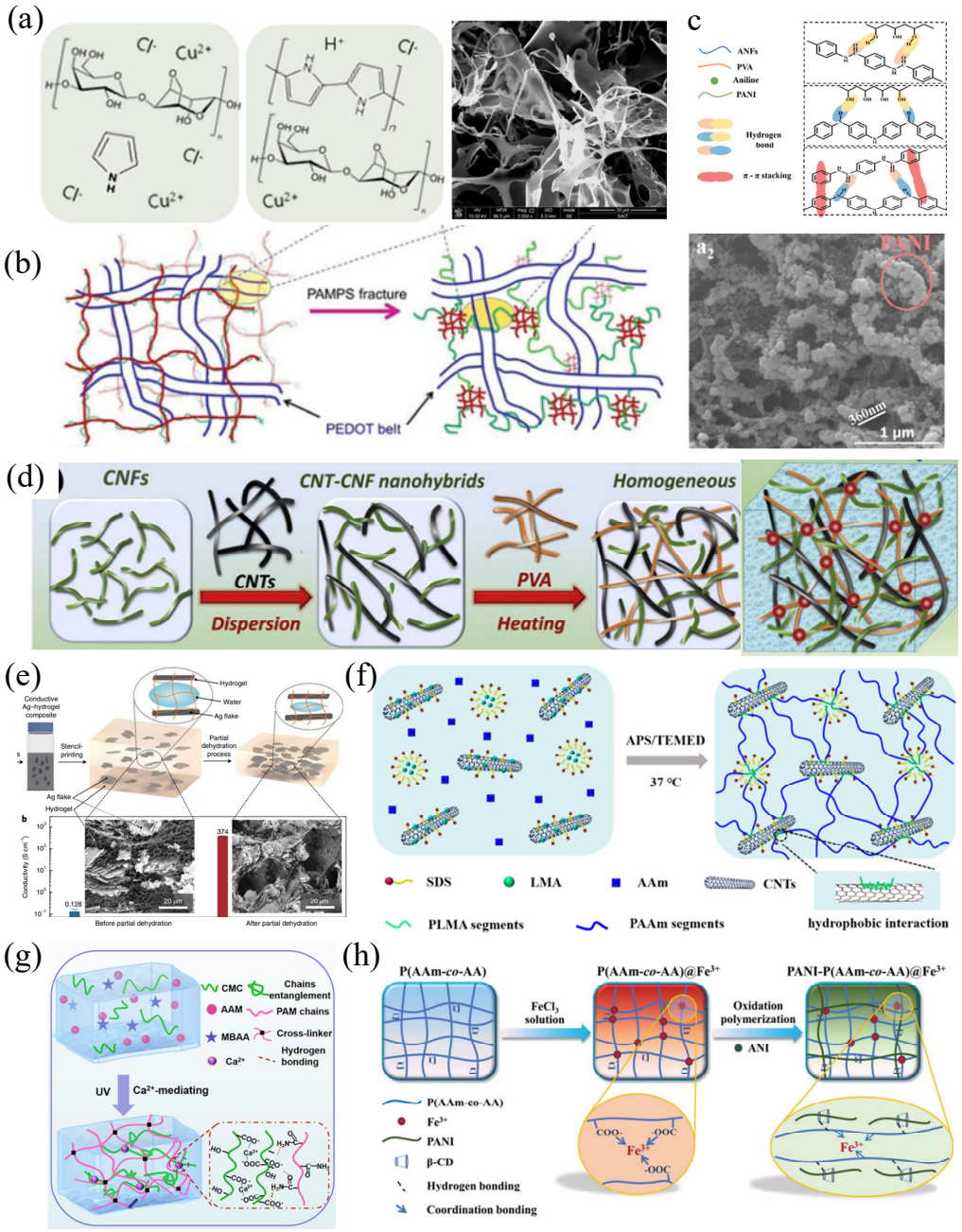


**Figure 2.** The three main categories of conductive hydrogels: Electronic conductive hydrogels: (a) polypyrrole (PPy) [24], (b) poly(3,4-ethylenedioxythiophene) (PEDOT) [25], and (c) polyaniline (PANI) [31]. Nanoparticle conductive hydrogels: including but not limited to (d) graphene [38], (e) carbon nanotubes [50] and (f) metal particles (e.g., silver flakes) [51]. Ionic conductive hydrogels: (g)and (h) consisting of various metal ions [23,49].

## 3. Key properties and enhancement strategies of conductive hydrogels

Achieving excellent electrical conductivity and effective current conduction are fundamental properties to fulfill. However, considering the diverse potentials of conductive hydrogels, it is often required to possess various desirable characteristics, thereby expanding the application scope and fields of these materials. The ion conduction of hydrogel follows the non-Faradaic process and

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different Ca2+ concentration will significantly change the conductivity (Figure 3a–c). These include but are not limited to: robust mechanical performance, sensitive mechanical responsiveness, long-term stability, and biocompatibility, among other multifunctional properties. Robust mechanical performance ensures that conductive hydrogels exhibit ideal toughness during stretching or compression. Long-term stability ensures that conductive hydrogels can recover on their own without external assistance, thus improving the hydrogel's lifespan and reducing costs. Biocompatibility ensures that drug-carrying electrically active hydrogels or the hydrogels themselves bond well with skin or body cells, reducing cellular toxicity. The design of these properties will broaden the application scope and fields of these materials, meeting the diverse requirements of conductive hydrogels in different environments such as smart wearable device applications and tissue engineering applications. This section discusses the design strategies for the aforementioned properties, aiming to provide readers with some insights.

*3.1 Conductivity*

Conductivity stands as the paramount property of conductive hydrogels. Coupled with the myriad other unique characteristics of hydrogels [52,53], conductivity further broadens the applications of hydrogels in tissue engineering [54,55], flexible electronics [56,57], and beyond. Enhancing conductivity constitutes a significant aspect of research in conductive hydrogel systems. There exist numerous methods to enhance the conductivity of conductive hydrogels. Highly conductive hydrogels typically achieve conductivity through ions or conductive polymers. Salt ions (ionic conduction) [58] or electrons within conductive polymers [59] undergo directed movement within hydrogels, thus imparting higher conductivity. Conductive properties of some typical conductive hydrogels are listed in Table 1. Zhou et al.'s study on conductive hydrogels incorporated NaCl salt ions to enhance conductivity on the basis of hydroxypropyl cellulose fiber conductivity [60]. Liu et al. [61] soaked methacrylic acid shrinkage glycerol ester (GSP) hydrogel in a 5 wt% NaCl solution, resulting in GSP-Na ion-conductive hydrogel. The mentioned gels, through the free movement of Na+ and Cl– ions within the hydrogel, embed the electrolyte into the entire polymer network, ensuring the continuity of the conductive phase throughout the deformation process. Increasing the concentration of salt ions in hydrogels can enhance charge transfer efficiency, further improving conductivity, thus rendering hydrogels with excellent conductivity [62]. Simultaneously, adjusting the ion content in hydrogel networks by varying the concentration and soaking time of NaCl solution can also regulate the hydrogel's sensitivity. Introducing conductive polymers (such as polythiophene, polypyrene, polybenzoxazole, polyaniline, etc.) as a second network on the basis of existing hydrogels, by increasing the number of conductive polymers, can enhance their conductivity. Researchers have also prepared meth acryloyl gelatin (GelMA) hydrogels encapsulating dorsal root ganglion (DRG) cells, integrating them with 3D printed conductive structures [63]. To enhance conductivity, they mixed a solution of PEDOT freeze-dried: polystyrene sulfonate (PSS) with polyethylene glycol diacrylate (PEGDA) as the basis for photocurable polymers, as illustrated in Figure 3d. The addition of PEDOT: PSS solution significantly increased the hydrogel conductivity, and with the increase in solution concentration, conductivity further improved (Figure 3e,f). For nanoparticle-based conductive hydrogels, Wang et al. [64] developed a fully natural ionic conductive hydrogel utilizing bentonite (BT) nanosheets and LiCl. According to zeta potential measurements, the cellulose/BT nanocomposite material carried a negative charge, and the gaps between adjacent BT nanosheets separated by cellulose served as channels for rapid cation transport, successfully achieving a conductivity of 89.9 mS/cm. Yao et al. [65] combined cellulose nanofibers (CNF) with PAM/phenylboronic acid-ionic liquid (PBA-IL) crosslinked networks to prepare multifunctional conductive hydrogels. CNF possesses a unique nanoscale structure capable of enhancing conductivity. PBA-IL and CNF synergistically enhance gel conductivity, improving ion transfer efficiency by generating multiple conductive pathways.

Sensitivity is also a pivotal attribute of conductive hydrogels. When subjected to external stimuli such as strain, pressure, temperature, humidity, or torsional deformation, conductive hydrogels undergo shape deformation, leading to changes in measurable electrical properties such as current,

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resistance, capacitance, among others, due to alterations in electron or ion transport pathways. Among various sensory properties, strain sensitivity is indispensable and has found widespread applications in conductive hydrogel systems. This variability in sensitivity can be characterized by the strain coefficient (GF) ($GF = (R - R_0)/R_0)/e$), where represents strain, and $R_0$ and $R$ denote the resistance at 0% strain and tensile strain, respectively [66,67]. A dual-conductive network hydrogel was developed (Figure 3g), merging the stretchability of double-network hydrogels with the conductivity of conductive nanowire films [68]. The resulting hydrogel sensor demonstrates ultra-sensitivity (GF = 343) and a broad sensing range (0-110 % strain) (Figure 3h). A simple transfer printing process incorporating carbon nanotubes introduces the conductive nanowire film into the hydrogel, facilitating the monitoring of various human activities such as finger bending, elbow bending, and knee movement. Ca2+ was incorporated into the crosslinked network of PAM-carboxymethyl cellulose sodium (CMCNa) to produce a transparent ionic conductive hydrogel [49]. With an increase in Ca2+ ion content, the efficiency of ion passage improves, leading to the formation of efficient ion transport channels or the generation of multiple conductive pathways, resulting in reduced resistance and increased conductivity. This effectively enhances the conductivity and sensitivity of the ionic conductive hydrogel. TA@talc particles were integrated into elastic PVA hydrogels, yielding an ionic conductive polyvinyl alcohol-tannic acid@talc supramolecular organic hydrogel [69]. Due to the uniform dispersion of ions, the prepared hydrogel exhibits strain sensitivity ($GF$ = 9.17, with a strain range of 0-1.2%), enabling it to function as a hydrogel-based sensor for detecting limb movements, pulse, speech, and handwriting. Additionally, it serves as a biological electrode for collecting electromyography (EMG) signals.

**Table 1.** Summary of conductive properties of some typical conductive hydrogels.

| | Conductive components | Conductivity (S $cm^{-1}$) | REF |
|---|---|---|---|
| Electronic conductive hydrogels | PEDOT: PSS | 10-2-4.38×103 | [70,71] |
| | PPY | 10-2-7.5×103 | [72–76] |
| | PANI | 10-2-2×102 | [77–79] |
| Nanoparticle conductive hydrogels | K+, Li+ | 0.02-0.0736 | [80,81] |
| | Ca2+ | 0.0337 | [19] |
| | Fe3+ | 0.00216 | [82] |
| Ion conductive hydrogels | CNTs | 0.082 | [83] |
| | MXene | 0.01092 | [84] |
| | Silver | ＞350 | [51] |

*3.2 Mechanical strength and flexibility*

Mechanical strength is also a crucial property of conductive hydrogels. In practical applications, conductive hydrogels require a certain level of mechanical robustness [85,86]. Conductive hydrogels based on macromolecules are often brittle and have limited ability to withstand mechanical loads. Therefore, to enhance the applicability and durability of conductive hydrogels, suitable parameters such as Young's modulus, high toughness, high fracture strength, and strain are essential mechanical properties for conductive hydrogels to withstand significant deformations under external forces. These parameters determine the application scope of hydrogels in tissue engineering and smart wearable devices. For conductive hydrogels, common strategies to improve their mechanical properties include double networks and nanocomposite networks.

Dual-network (DN) conductive hydrogels consist of a rigid, brittle polymer network as the first network and a flexible, tough polymer network as the second network [87]. The rigid network is typically crosslinked by covalent bonds, which act as sacrificial bonds, dissipating a significant amount of energy during large deformations, thereby maintaining the plasticity of the polymer network and preserving the integrity of the hydrogel [69]. This approach addresses the poor

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mechanical performance and lack of rapid self-recovery and self-healing mechanisms found in conventional single-network ion-conductive hydrogels [88–90]. For ion-conductive hydrogels containing conductive polymers, the conductive polymer chains typically serve the function of the first network, while the second network is often composed of flexible polymer chains such as PVA and PAM. For instance, Wang et al. [91] reported dual-network conductive hydrogels where they incorporated polyaniline (PANI) into a poly (acrylamide-co-hydroxyethyl methacrylate) (P(AM-co-HEMA)) network via in situ polymerization. Due to the inherent interactions between the conductive PANI network and the flexible P(AM-co-HEMA), the prepared hydrogels exhibited excellent strength and toughness under cyclic loading. With an increase in the degree of PANI oxidation, both the mechanical properties and electrical conductivity of the hydrogels were enhanced. Additionally, soaking in salt solutions can enhance the electrical and mechanical properties of DN conductive hydrogels, as they are mutually influential. In the conductive hydrogels prepared by Zhou et al., hydroxypropyl cellulose (HPC) biopolymer fibers were physically crosslinked with a tough and biocompatible PVA gel matrix, followed by immersion in a salt solution to obtain ion-conductive hydrogels [60]. The increase in HPC content resulted in higher tensile strength and stress of the conductive hydrogels, accompanied by an increase in electrical conductivity. This is attributed to the increased number of pores within the hydrogel, providing channels for the migration of salt ions, thus enhancing conductivity. Wei et al. [92] utilized a method involving PVA pre-gels immersed in (NH4)2SO4 solution. This process induced PVA chain folding and precipitation due to the Hofmeister effect, resulting in ion-conductive hydrogels with a tensile strength of 4.1 MPa. In another study, dual-network ion-conductive hydrogels were created, incorporating sodium caseinate (SC) and PAM/carboxymethyl chitosan sodium (CMS-Na) [93]. These hydrogels showcased outstanding mechanical properties and flexibility, utilizing the natural solubility of SC and its ability to spontaneously form micelle structures in water as energy dissipation centers. The in-situ synthesis of Ag/TA@CNCs within polyvinyl alcohol hydrogels to fabricate multifunctional nanocomposite hydrogels [32] (Figure 3i). These nanohybrids, such as LAPONITEs XLS (clay nanosheets), possess hydrophilicity and can be utilized as Nano-reinforcement materials and physical crosslinkers for synthesizing nanocomposite hydrogels with high tensile strength. This significantly enhances the hydrogel's elastic modulus, toughness, and extensibility (Figure 3j,k). Additionally, ion-conductive dual-network hydrogels were achieved by immersing polyacrylamide/gelatin networks in sodium citrate solution [94] (Figure 3l). The ion crosslinking in the gelatin network served as reversible sacrificial bonds, absorbing energy during stretching and quickly reforming, thereby resulting in hydrogels with high mechanical performance (Figure 3m,n). The network of nanocomposites is crucial in the realm of conductive hydrogels. This network can consist of exfoliated clay nanoparticles or simple fillers mixed within polymer matrices (such as carbon nanotubes, reduced graphene oxide, etc.). The uniform and continuous three-dimensional mesh structure grants hydrogels inherent flexibility and excellent mechanical properties. While conductive fillers in conductive hydrogels indeed enhance their electrical conductivity, their aggregation tends to disrupt the uniformity of the hydrogel structure, thereby compromising its mechanical performance. Incorporating conductive fillers into hydrophilic nanomaterials or modifying hydrophilic nanomaterials can significantly enhance the dispersibility of conductive materials. Consequently, incorporating nanocomposite materials into hydrogels can markedly improve their mechanical properties. For instance, incorporating oxidized multi-walled carbon nanotubes (oxCNTs) into polyacrylamide (PAM) hydrogel matrices enables the development of conductive nanocomposite hydrogels [95]. By functionalizing oxCNTs, aggregation within the hydrogel network is circumvented, effectively enhancing the material's mechanical properties. The resulting hydrogel exhibits exceptional tensile performance and recyclability. Tondera and colleagues [96] demonstrated that the synergistic interaction of nanoclay colloid dispersion with PEDOT and secondary polymer networks of polyacrylamide results in multi-network hydrogels with high electrical conductivity, stretchability, and printability. The excellent flexibility also relies on the intermolecular interactions within the hydrogel. Researchers have devised an exceptionally stretchable hydrogel by harnessing tannic acid-activated dynamic interactions (TEDIs) [97], replacing conventional covalent cross-linking. The TEDI

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hydrogel exhibits a remarkable stretching capability exceeding 7300 %, which is 40 times that of chemically cross-linked hydrogels. Its potential in accurately detecting human motion for wearable electronic devices and healthcare monitoring, thus holding considerable application value. Additionally, Zhang et al. [98] employed a one-pot synthesis method to develop a novel amphiphilic ionic liquid polymer hydrogel. By introducing acrylamide into the hydrogel composed of zwitterionic liquid and methacrylic acid lysine monomers, the hydrogel's strength is enhanced. The introduction of ZIL and lysine functional groups facilitates numerous non-covalent interactions (hydrogen bonds and ionic bonds) within the hydrogel. The outstanding mechanical properties of the Am-ZIL-LysMA hydrogel, with stretching exceeding 2000 %, demonstrate excellent stability, repeatability, and high strain sensitivity across a wide operating range.

Given the above considerations, different applications have varying requirements for the mechanical and electrical properties of conductive hydrogels. Conductive hydrogels used in electronic components demand high mechanical conductivity and electrical conductivity. Conversely, those utilized in sensors may not require high electrical conductivity but place significant emphasis on properties such as stretchability, compressibility, and flexibility. Therefore, in practical applications, there needs to be a balance between mechanical conductivity and electrical conductivity, tailored to the specific requirements of each application.

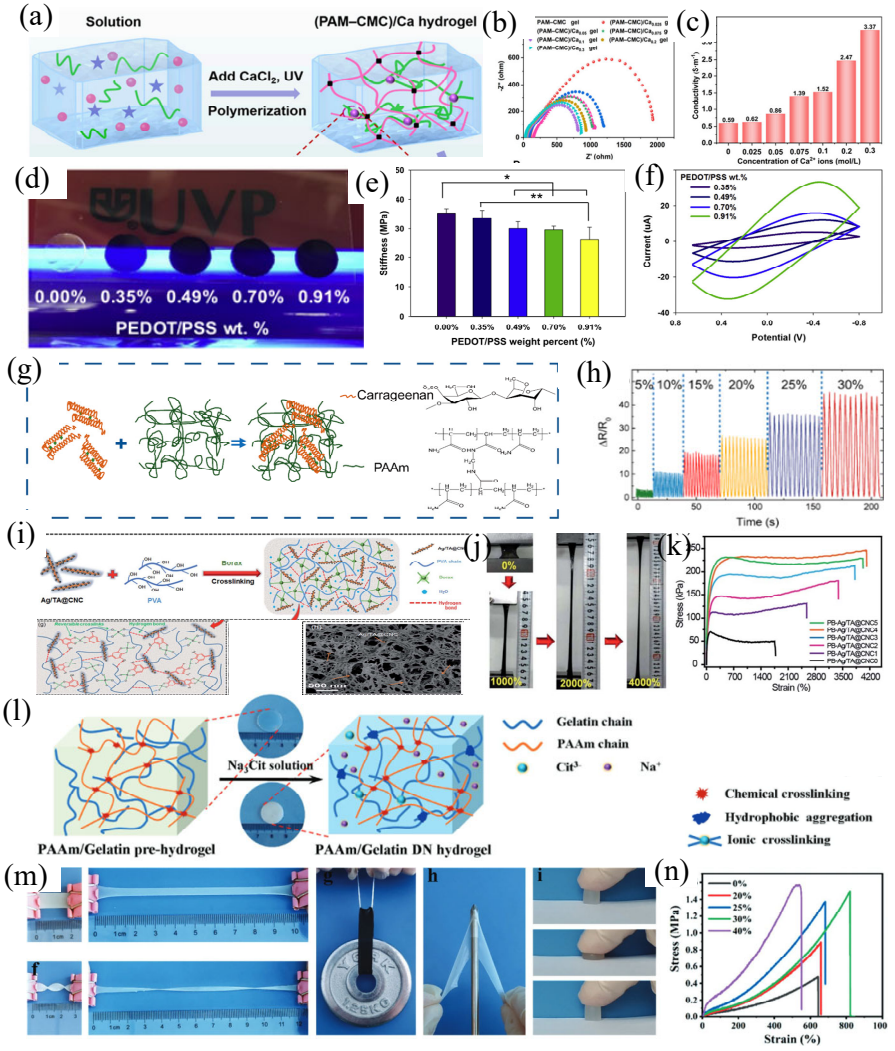


**Figure 3.** Conductivity and mechanical properties of conductive hydrogels: (a) Fabrication of (PAM–CMC)/Ca hydrogel for iontronic sensors. (b) the EIS Nyquist plots and (c) the conductivity of (PAM–CMC)/Ca hydrogel with different Ca2+ content [19]. (d) conductive hydrogels with different concentrations of PEDOT/PSS. (e) Mechanical properties and (f) Electrochemical properties CV of

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conductivity hydrogel with different PEDOT/PSS content with scan range from −0.8 to 0.65 V [63]. (g) the network structure of the carrageenan/PAAm hydrogel. (h) Stable signal output under different strains [68]. (i) Chemical cross-linking reaction in PB-Ag/TA@CNC hydrogels. (j) and (k) Excellent stretchable and mechanical properties of PB-Ag/TA@CNC4 hydrogels [32]. (l) illustration of PAAm-Gelatin DN hydrogel. (m) and (n) Excellent stretchable and mechanical properties of AAm-Gelatin DN hydrogel [94].

*3.3 Long-term stability*

With the widespread utilization of conductive hydrogels across various domains, particularly in flexible wearable devices for the human body and tissue engineering, there is an imperative for them to possess extended operational lifespans. Furthermore, the incorporation of healing properties in flexible electronic materials can significantly prolong the lifespan of flexible wearable electronic devices, achieving the objectives of resource conservation and cost reduction. Consequently, the long-term stability of conductive hydrogels poses a significant challenge. In recent years, extensive research has been conducted on the self-healing and water retention properties of conductive hydrogels, with these outstanding attributes greatly enhancing their long-term stability.

Self-healing of hydrogels can also be achieved through molecular self-assembly. Wang et al. [110] utilized nucleoside monomer molecules (2-FA) and distilled water/phosphate-buffered saline (PBS) as solvents to prepare a high-strength, injectable supramolecular hydrogel by constructing a multi-hydrogen bond system. Due to the hydrogen bonds and n-π stacking interactions between molecules in the self-assembled system, the 2-FA hydrogel exhibits excellent self-healing and shear-thinning properties, transitioning from a liquid to a gel state within seconds after injection at 37°C. Exposure to working environments for extended periods can lead to dehydration and dryness, limiting the working environment and lifespan of conductive hydrogels. Therefore, the hydrogel's moisture retention is also one of the key characteristics determining its long-term stability. Some studies involve adding coatings to the surface of hydrogels to physically reduce water evaporation rates [111]. Currently, the main methods to enhance the moisture retention of ion-conductive hydrogels include the following: 1) encapsulating ion-conductive hydrogels with elastic materials such as polyethylene films, polydimethylsiloxane (PDMS), or Ecoflex to form a sandwich structure of elastic material-hydrogel-elastic material; 2) introducing highly hygroscopic salts or alcohols into the hydrogel system; 3) incorporating other moisture-retaining substances (such as zwitterions, silk fibroin) into the ion-conductive hydrogel system [66,83,112,113]. While encapsulating with elastic materials improves the moisture retention of ion-conductive hydrogels, it reduces their sensitivity and adhesion. Introducing alcohol substances, highly hygroscopic salts, or other materials with special moisture-retaining structures can effectively enhance the moisture retention of ion-conductive hydrogels. Wang et al. [114] encapsulated the prepared ion-conductive hydrogel with polyethylene film and placed it between two aluminum electrodes to prepare a pressure sensor with long-term stability (Figure 4a,b). Wu et al. [115] immersed PAM/carrageenan double network hydrogels in a LiBr aqueous solution. With the ionization of Li+ and Br– in the solution permeating into the hydrogel due to concentration gradients, the ion-conductive hydrogel with moisture retention and antifreeze properties was obtained. The introduction of LiBr enhances its stability and working temperature range, allowing the ion-conductive hydrogel to exhibit a super-high elongation rate (625 % strain) at -78.5°C, indicating its potential as a high-performance sensor even in harsh environments. Yang et al. [116] incorporated silk fibroin (SF) into PVA and boric acid (Bx) to prepare ion-conductive hydrogels with good moisture retention. SF with its unique β-folded microstructure contributes to improving the moisture retention of the hydrogel. The inclusion of SF resulted in the hydrogel retaining 75 % of its moisture after being placed at room temperature for 2 days and 60 % after 4 days. Organic solvents (such as ethylene glycol, glycerol), ionic compounds, and zwitterions (such as betaine, proline) can bind water molecules to synthesize mixed organic hydrogels with a wide working temperature range [117]. Fibers of poly (N-acryloyl glycinamide-co-acrylamide) (PNA) hydrogels prepared by Shuai et al. [118] exhibit high tensile strength, excellent extensibility, good conductivity, and self-healing capability. Coating elastic poly (methyl acrylate) (PMA) on PNA hydrogels enhances their moisture

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retention. He and colleagues [119] developed a freezing-resistant moisturizing hydrogel by combining TA-CNT nanocomposites with water-glycerol dispersion media in a polyvinyl alcohol matrix (Figure 4e). Compared to hydrogels without water-glycerol dispersion media, the prepared hydrogel exhibited excellent antifreeze properties (-30 °C) and long-term moisturizing performance (10 days). Han and colleagues [83] incorporated polydopamine (PDA)-modified carbon nanotubes into hydrogels composed of water and glycerol binary solvent systems (Figure 4f,g). The prepared hydrogel exhibits heat resistance and maintains its performance over a wide temperature range (20 ~ 60 °C) and for a relatively long time (30 days) under room temperature conditions (25 °C).

The primary focus lies in the exploration of self-healing capabilities. Traditional conductive hydrogels are largely disposable, losing their utility upon rupture. With the growing emphasis on sustainable development, the demand for durable materials has surged, making the design of conductive hydrogels with self-healing properties increasingly crucial. Drawing inspiration from the observed self-repair abilities of organisms following mechanical injuries, the integration of self-repair characteristics into hydrogels significantly enhances their durability and stability. Self-healing harnesses the dynamic properties of hydrogels to restore their structure and functionality post-injury, thereby prolonging their lifespan. Zhang et al. [106] prepared multifunctional ion-conductive hydrogels using PAA, DA-functionalized hyaluronic acid (DHA), and Fe as raw materials, exhibiting excellent autonomous healing performance (as shown in Figure 4c). Due to the rich hydrogen bonding and various metal coordination interactions between Fe, catechol, and -COOH, the hydrogel demonstrates outstanding self-healing performance in both mechanical and electrical properties (98 % recovery within 2 seconds). Additionally, this advancement has spurred innovative applications, leading to a burgeoning interest in self-repair hydrogels [21]. The self-healing properties of conductive hydrogels primarily stem from the dynamic bonding and interactions present within the hydrogel matrix (Figure 4d) [99]. Dynamic covalent bonds refer to chemical bonds that can reversibly break under mild conditions. Common forms of dynamic covalent interactions include imine bonds, acyl hydrazone bonds, disulfide bonds, boronic ester bonds, and Diels-Alder reactions. Under gentle conditions, damage can be repaired through the reconstruction of dynamic covalent bonds, endowing the hydrogel with self-healing capabilities [100,101]. Typically, the self-healing process of ion-conductive hydrogels requires external stimuli [102] (such as temperature, pH, and light). Peng et al. [103] fabricated a novel thermo-plastic ion-conductive hydrogel based on PVA/NaC1/glycerol, exhibiting thermal healing capacity and thermo-plasticity at 120°C. The presence of hydrogen bonds between glycerol and PVA chains is a critical factor contributing to the thermal healing capacity and thermo-plasticity of the hydrogel, resulting in strain and stress self-healing efficiencies of 70 % and 95 %, respectively. Furthermore, Hua et al. [104] employed synergistic multiple interactions between montmorillonite (MMT), poly(acrylamide-co-acrylonitrile) (P(AAm-co-AN)), xanthan gum (XG), and Fe3+ to develop XG/MMT/PAAm hydrogels with toughness, conductivity, and self-healing properties. Currently, various self-healing ion-conductive hydrogels based on reversible non-covalent interactions have been developed, eliminating the need for external stimuli during the self-healing process. Du's research team [105] utilized poly(aniline) (PANI) and polyurethane (PU) as raw materials to fabricate self-healing conductive hydrogels through reversible Diels-Alder reactions. After 2 hours of storage at room temperature without external stimuli, the hydrogel's fracture elongation and electrical properties could recover by 65.4% and 90%, respectively, demonstrating the broader application of Diels-Alder reactions in the field of self-healing.

Compared to dynamic covalent bonds, dynamic non-covalent bonds, due to their weaker bond strength and greater reversibility, are more suitable for fabricating self-healing conductive hydrogels. Zhao et al. [103] utilized Bx as a cross-linking agent to prepare a multifunctional self-healing ion-conductive hydrogel. Due to the abundance of borate ester bonds and hydrogen bonds within the hydrogel network, its self-healing efficiency reached as high as 93.56 % without external stimuli, demonstrating excellent sensing performance post-healing. With this dynamic cross-linking network, the composite hydrogel can reshape into various forms under external pressure. The borate salts and hydroxyls on adjacent PVA chains are prone to break and recombine, thus completing the self-repair process in a short timeframe. In a separate study, researchers [107] developed a novel multifunctional

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conductive polymer hydrogel using multiple hydrogen bonds to address the issue of inadequate mechanical properties of hydrogels. Results showed that the hydrogel's electrical conductivity could restore to its original state after multiple cutting cycles. The reported hydrogel could even regain its original strength despite severe damage (1000 % strain), owing to the dynamic dissociation/recombination process of hydrogen bonds within the hydrogel. In another approach, scientists [16] incorporated carbon nanotubes (CNTs) into a chelate of calcium ions (Ca2+), polyacrylic acid (PAA), and sodium alginate (SA), designing a bio-inspired (Ca-PAA-SA-CNTs) hydrogel with multiple conductivity capabilities. Additionally, printing the hydrogel onto a stretchable substrate and connecting it with electrodes resulted in a strain sensor resembling human skin. This sensor exhibited sensitivity to external stimuli such as finger bending, knee bending, and respiration, offering stable responses in the form of relative resistance and relative capacitance changes, thereby providing new avenues for exploring the application of advanced hydrogels in multifunctional smart wearable devices. Tie et al. developed a high-strength, compressible, self-repairing tactile platform by mixing Fe3+ ions with PVAA/PAM hydrogel [108]. Due to the rapid chelation of Fe3+ with the acyl dichloride on the PVAA chains, the hydrogel exhibited excellent self-healing properties (80 % recovery within 24 hours). Ren et al. synthesized a self-healing hydrogel [109] using o-carboxymethyl chitosan (O-CMCS) and PVA as raw materials. Due to the complexation of poly-b-cyclodextrin with diamond and the interaction between dynamic Schiff bases and borate ester bonds, the OPPC hydrogel demonstrated good conductivity, excellent mechanical strength, and self-healing properties, with a self-healing efficiency exceeding 95 % within 15 seconds. Thus, dynamic non-covalent interactions offer numerous options for manufacturing self-repairing conductive hydrogels with different polymer networks.

*3.4 Biocompatibility*

Biocompatibility typically refers to the ability of biomaterials to induce appropriate host responses in biological systems. For conductive hydrogels used in applications such as human tissue engineering and wearable flexible devices, biocompatibility is crucial as they come into direct contact with biological interfaces. There are many factors influencing biocompatibility, such as chemical composition, surface charge, surface morphology, pH, and others. Although commonly used materials like gold and steel are inherently non-toxic, they often provoke inflammation due to immune reactions after implantation, which can pose a threat to the host's life. Therefore, various methods are being employed to improve the biocompatibility of hydrogels, aiming to develop materials that can interact with the body in a more targeted and controlled manner.

Currently, materials with excellent biocompatibility mainly come from natural sources, including plants and animals. Examples of such materials include chitosan [120], alginates [121], cellulose [122], agarose [123], as well as synthetic polymers and inorganic substances (bioceramics [124], titanium alloys [125]). Utilizing natural materials has several advantages. Firstly, they are generally available at lower costs. Secondly, these materials possess molecular structures similar or identical to those found in the human body, thereby minimizing the likelihood of triggering severe immune reactions. Finally, natural materials are easily biodegradable in the body, resulting in the production of small, less biotoxic molecules after degradation. The pre-gel synthesized from CS and AM was immersed in a concentrated NaCl solution, resulting in the formation of ion-conductive hydrogels characterized by cell compatibility and antibacterial properties, which stem from the inherent traits of amine-rich polymers and a straightforward synthesis approach [126]. Following 24 hours of in vitro cytotoxicity testing, cell viability stood at 98%, and over a span of 6 months in the laboratory, no microbial growth was detected. In another study, a biocompatible ion-conductive hydrogel was prepared through the collaborative coordination of tannic acid (TA)-coated cellulose nanocrystals (CNC), PAA chains, and Al3+ ions [127]. The incorporation of catechol groups from TA facilitated direct adhesion of the hydrogel to human skin, bypassing inflammation concerns (Figure 4j). Furthermore, the development of a conductive polymer hydrogel (CPHs) utilizing tannic acid (TA) and polypyrrole (PPy) as crosslinkers demonstrated superior electrochemical impedance and conductivity compared to conventional PPy hydrogels [128]. Upon degradation into smaller particles

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within the spinal cord, CPHs prompted a diminished initial inflammatory response, underscoring their excellent biocompatibility (Figure 4h,i). Moreover, integration of N-(3-Dimethylaminopropyl)-N'-ethyl carbodiimide hydrochloride into a crosslinking strategy, alongside poly (3,4-ethylenedioxythiophene) – poly (styrene sulfonate) and carbonylated multi-walled carbon nanotubes, enabled periodic electrostimulation of the MESGel scaffold co-cultured with hamster lung cells [16]. The observed decrease in dead cells after 5 days of MESGel electrostimulation significantly facilitated wound healing, highlighting the extensive potential applications of this conductive hydrogel in the biomedical domain.

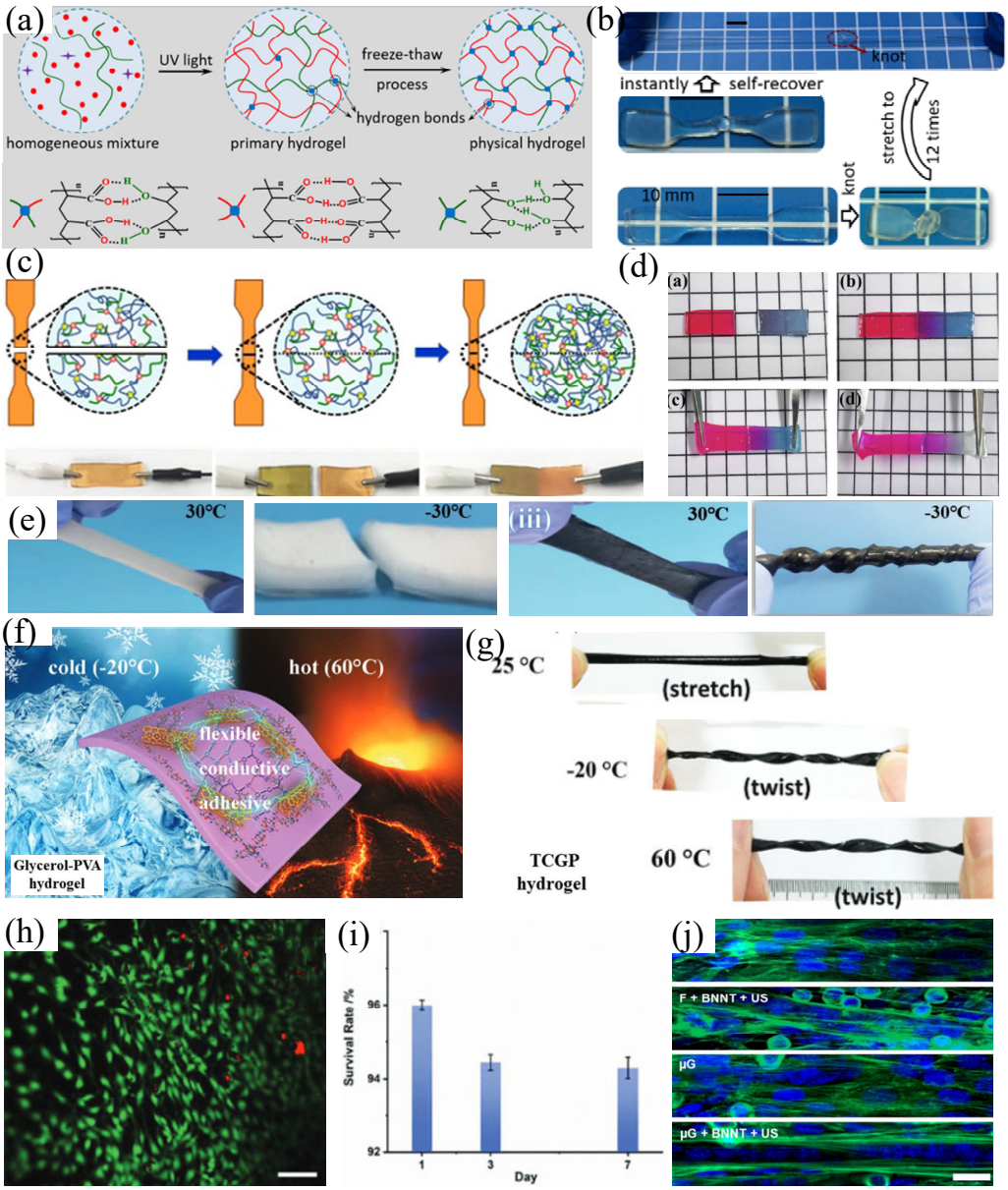


**Figure 4.** Stability and biocompatibility of conductive hydrogels: (a) Preparation process of SPP hydrogel. (b) Hydrogen bonding dominates good self-healing properties [114]. (c) Illustration of the self-healing mechanism of the PAA/CMCx–Fe3+–S hydrogels, ionic dynamic bonds dominate good self-healing properties [106]. (d) H-A-P hydrogels, mainly hydrogen bond dominated self-healing [99]. (e) and (f) GW hydrogel shows antifreeze and high temperature resistance (-20℃-60℃) [83]. (g) The freezing resistances of glycerol–PVA and TCGP hydrogels [119]. (h) Fluorescence image of cells cultured on conductive hydrogel film. Live cells appear green, while dead cells appear red. (i) Cell survival rate on conductive hydrogel film on days 1, 3 and 7 [128]. (j) Differentiation of skeletal muscle cells on conductive hydrogel [127].

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## 4. Application demonstration in flexible electronics

As a novel soft material, multifunctional conductive hydrogels have garnered increasing attention due to their flexibility, fatigue resistance, excellent electrical properties, tunable mechanical performance, stability in different environments, and "smart" capabilities associated with different components. They have already found applications in wearable sensors, medical monitoring devices, tissue engineering, and more [129]. Functioning as a flexible, highly stretchable, adhesive, biocompatible, and antibacterial conductive material, ionically conductive hydrogels demonstrate enormous potential in flexible sensors and biomedical applications [130–132]. Their versatile properties make them promising candidates for various fields, offering solutions to diverse challenges in flexible electronics and healthcare.

### *4.1 Applications in tissue engineering*

Modern scientific research confirms that the extracellular matrix (ECM) is a hydrated network composed of biomacromolecules, providing biochemical and structural support to cells. Replicating the natural ECM with chemically defined synthetic polymers has long been a challenge in cell culture and tissue engineering. Hydrogel scaffolds can achieve desired ECM-like effects through design and programming, providing support similar to the ECM. It has been extensively demonstrated that hydrogels can promote cell attachment, proliferation, and differentiation, while electrical signals control the metabolism, adhesion, proliferation, migration, and differentiation of electrically excitable cells [133]. Additionally, many cell types, such as epithelial cells, fibroblasts, and cardiomyocytes, are sensitive to material stiffness [134–137]. Therefore, the conductivity and mechanical properties of conductive hydrogels will significantly influence cell behavior, leading to promising applications in tissue engineering.

In recent years, significant progress has been made in neural tissue engineering for nerve injury repair and neural function regeneration [138]. Among various scaffold materials, hydrogels are ideal for neural repair due to their good biocompatibility, high water content, and mechanical properties similar to natural neural tissue. Neural information transmission relies on electrical signals [139]. Due to the excellent conductivity of conductive hydrogels, they can assist in recording electrical signals induced by electrical stimulation. Moreover, hydrogels can also transport drugs/cells to target sites [140,141]. On the other hand, electrical signals are crucial for neuronal cell survival, differentiation, and activation of neural circuits. Studies have shown that electrically conductive hydrogels hold great potential in neural tissue repair [136–138].

A positively charged nerve conduit was developed using oligo (polyethylene glycol) fumarate (OPF) hydrogel [142]. Enhanced proliferation and diffusion of PC12 cells on conductive hydrogels in a cell matrix-like environment were demonstrated. A dynamic conductive hydrogel (DCH) composed of collagen, alginate, and PEDOT: PSS was reported [143]. This material mimicked the fibrous structure of the ECM, offering desired stiffness and stress relaxation properties. It significantly improved electrical coupling in engineered cardiac tissues, enhancing beating characteristics. Fabrication of a DCH consisting of gelatin meth acryloyl (GelMA) and PEDOT: PSS for bioprinting and constructing conductive 3D structures loaded with C2C12 myoblasts was carried out [144]. An ionically conductive hydrogel based on an ionic conductor basified (trifluoroethane) lithium was prepared [145]. A friction-based nanogenerator constructed from this hydrogel was successfully implanted into the patellar ligament of rabbit knees. Both in vitro and in vivo tests indicated no cytotoxicity and excellent biocompatibility. The hydrogel sensor converted mechanical energy into electrical energy, providing stable electrical output signals during repetitive stretching and bending cycles of rabbit legs at different frequencies (~0.4 Hz and 1.5 Hz) and different bending angles (~125° and 65°), potentially offering new solutions for real-time diagnosis of muscle and ligament injuries. A light-responsive and stretchable ECH was developed by copolymerizing polyaniline and polyacrylamide (PAM) [146]. Under near-infrared light irradiation, the conductivity of this ECH was enhanced, facilitating the conduction of bioelectric signals. In vivo studies showed that highly conductive bridging implants could replace the missing sciatic nerve, suggesting that ECH may be a potential biomaterial for replacing damaged peripheral nerves in tissue engineering

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(Figure 5a). A pioneering microneedle (MN) array patch integrated with carbon nanotubes (CNTs) was introduced for the treatment of myocardial infarction (MI) by facilitating the attachment and alignment of induced pluripotent stem cell (iPSC)-derived cardiomyocytes (CMs) [20] (Figure 5b). A silk-free electrospinning (ES) method for spinal cord regeneration was developed [147]. Magnetoelectric Fe3O4@BaTiO3 nps loaded with hyaluronic acid (HA)/collagen hydrogel were prepared, and induced ES using an external magnetic field promoted nerve regeneration in a rat hemi section spinal cord injury (SCI) model.

Incorporating antimicrobial quaternary ammonium compounds and curcumin, among other substances beneficial for wound healing, into conductive hydrogels presents a promising application in the field of biomedicine, both as wound dressings and for topical use on the skin. Skin, being the body's largest organ, directly interfaces with the external environment, serving a critical role in preventing the invasion of pathogens and microorganisms, thus playing a vital role in maintaining overall health. However, the skin is also a highly vulnerable area, with an increasing number of cases of skin defects resulting from external traumas, burns, ulcers, surgical wounds, and the like. Unlike traditional dressings that typically require medication for wound repair, electrically active hydrogels can facilitate wound healing through their inherent electrical properties. This eliminates the need for frequent dressing changes, reduces reliance on external medications [42], and alleviates concerns about potential over- or under-dosing of traditional dressings [148].

Skin is also sensitive to electrical signals, as epithelial tissues transport ions across the epidermis, functioning as an epidermal battery. When used as wound dressings, conductive hydrogels not only possess wound detection capabilities but also offer the advantage of directly stimulating cells to promote wound healing. In intact skin, there is a polarization distribution of ion channel charges in epithelial cells [21,149]. When tissue is disrupted, the balance of transmembrane potential is disturbed, resulting in a decrease in potential at the wound site. Studies have shown that electrically active dressings containing conductive polymers exhibit conductivity comparable to that of the skin (10-60 mV) and can assess the extent of wound recovery [150]. It is worth emphasizing that wound detection devices not only need to monitor changes in resistance and current but also require hydrogels to be flexible to ensure proper contact between the sensing electrodes and the wound tissue, thus obtaining accurate electrical signals [151,152].

Electrical stimulation can induce cell migration and proliferation along electrical gradients until wound closure, reconstructing the original epidermal battery's trans-epithelial potential (TEP) [153]. Conductive hydrogel dressings can provide both an electrical pathway for endogenous bioelectricity and guidance for cell adhesion, migration, and orientation, thereby facilitating wound healing. Researchers have synthesized a two-dimensional biocompatible nanosheet-based hydrogel using cellulose crystals and PDA-reduced graphene oxide. The assembled nanosheet hydrogel exhibits biological stability, conductivity, flexibility, and cell/tissue affinity. Additionally, Wang et al. prepared antimicrobial polyampholyte conductive hydrogels using SBMA and HEMA [154]. The abundant quaternary ammonium groups in the hydrogel interact with bacterial membranes, disrupting their structure and demonstrating over 99% antimicrobial efficacy, making them suitable for wound dressings. Furthermore, Shen et al. synthesized a nano-drug ion hydrogel using HLC-EPL, OHA, curcumin (CuR), and Fe3+ [155]. HLC-EPL and OHA, similar to skin components, are biodegradable substances with hemostatic, anti-inflammatory, nutritional support, and accelerated vascular formation properties, achieving a combined strategy for dressing-nanodrug therapy (Figure 5c). However, during wound detection, it is crucial to differentiate between the resistance of the dressing and that of the wound. The detected resistance should accurately reflect the wound resistance after excluding the resistance introduced by the dressing [156]. Additionally, in cases of bacterial infection, the formation of a biofilm on the wound can lead to reduced resistance, affecting the accuracy of wound detection. Ideally, preventing multiple wound infections is desirable; however, current electrically active wound dressings used for wound detection may not completely eliminate this issue.

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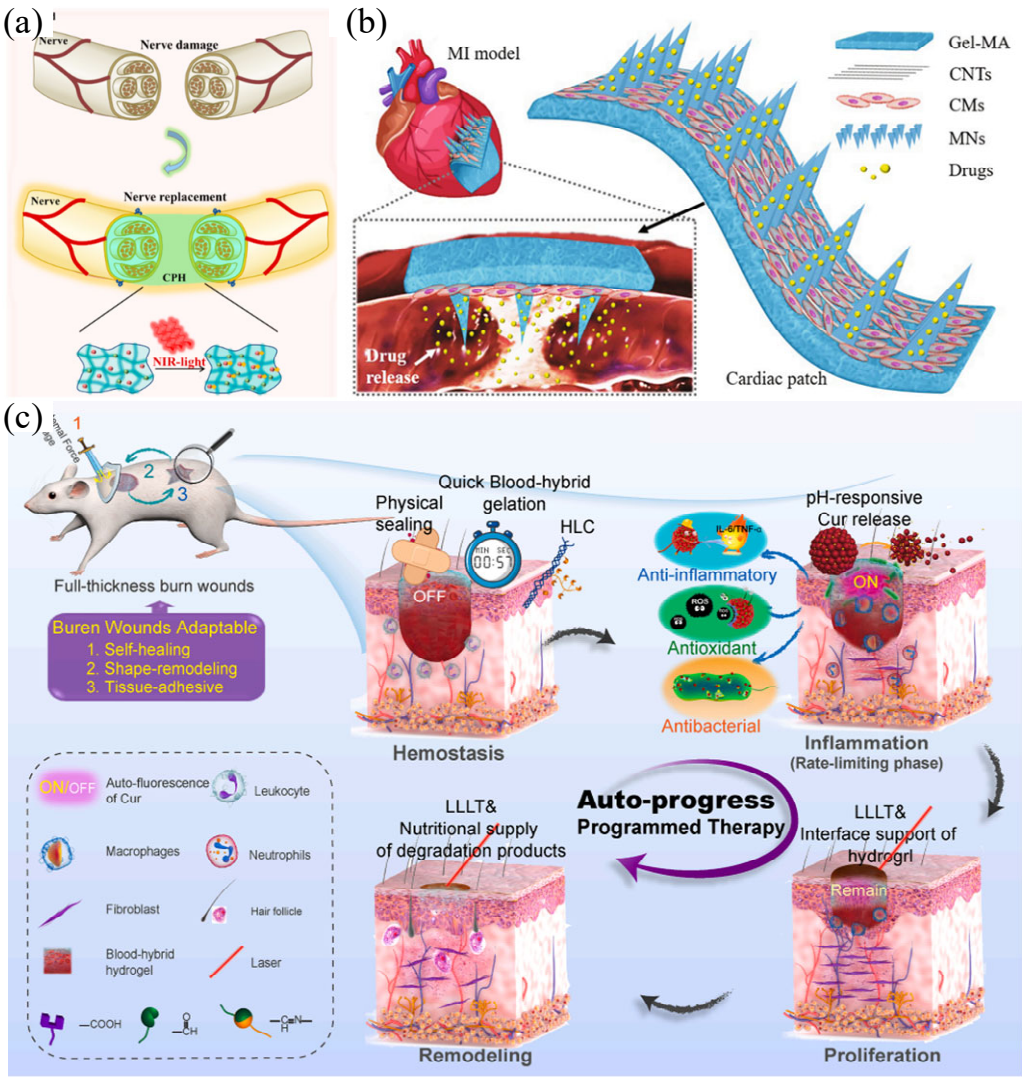


**Figure 5**. Several applications of conductive hydrogel in tissue engineering: (a) Conductive polymer hydrogel used to replace damaged nerve [146]. (b) Carbon nanotube conductive hydrogel is used to treat infarcted heart [20]. (c) Treatment process of cu - fe (III) - HEO nanocomposite hydrogel for burn wound dressing [155].

*4.2 Applications in soft wearable devices*

Wearable multifunctional sensors serve as interfaces between human soft tissues and electronic devices, holding vast potential in signal acquisition, drug delivery, and beyond [157,158]. In recent years, with the rapid development of flexible electronic materials, wearable flexible sensors have shown promising applications in fields such as artificial intelligence, motion monitoring, medical rehabilitation, and soft robotics [159]. Wearable devices can detect both large deformations closely related to personal physiological conditions (such as joint movements) and small deformations (such as pulses and heartbeats). Therefore, wearable sensors play a crucial role in monitoring human health. Flexible sensors are categorized into resistive sensors, capacitive sensors, friction-based sensors, and piezoelectric sensors, enabling continuous and real-time monitoring of human joint movements and physiological signals (such as heart rate, blood pressure, etc.) [160,161] (Figure 6a,b). Moreover, owing to the adhesive, flexible, conductive, and stable nature of ion-conductive hydrogels mimicking human skin, they can adhere to the surface of biological tissues without the need for additional media. This helps reduce signal noise during monitoring, ensuring both sensitivity and accuracy of flexible sensors while providing a certain level of wearing comfort. As a result, they have become powerful candidates for flexible wearable electronic materials.

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Due to the sensitivity of conductive hydrogels, they hold great promise for use in medical diagnostics as wearable devices. Integrating hydrogels onto the throat and chest can detect physiological movements such as speaking and different breathing patterns [34,91,107] (Figure 6c). Furthermore, the shape of pulses can be analyzed based on the frequency of output signals. By recording different pulse signals in various states and comparing them with healthy thresholds, one can observe a person's health condition. Additionally, electromyograms (EMGs), electrocardiograms (ECGs), and electroencephalograms (EEGs) are crucial probes for human electrophysiological signals, allowing the determination of the functional status of peripheral nerves, neurons, neuromuscular junctions, and muscles. Detection of electrophysiological signals can be achieved through conductive hydrogel electrodes.

The research conducted by multiple teams has led to significant advancements in ion-conductive hydrogels for wearable sensors. In one study, multifunctional cellulose-based hydrogels were developed using tannic acid (TA), glycerol, and NaCl [162]. These hydrogels enable real-time monitoring of respiration and diagnosis of obstructive sleep apnea syndrome by detecting temperature changes from nasal airflow and mechanical vibrations from various body parts independently. Another study focused on a multifunctional conductive hydrogel composed of carboxyl-functionalized multi-walled carbon nanotubes (c-MWCNTs) cross-linked with polyacrylamide (PAM)/chitosan (CS) hybrid networks [34]. This hydrogel responds to bending movements of different body joints and small gestures like smiling and speaking. It can adhere to the chest and wrist for monitoring respiration and pulse in various modes, aiding in human health monitoring. Moreover, researchers utilized biocompatible materials such as PVA, phytic acid (PA), and gelatin (Gel) to create ion-conductive hydrogels [163], which assemble into portable sensors with Bluetooth components. These sensors provide real-time monitoring of human electrophysiological signals, including clear heartbeat signals without the need for bulky accessory devices. Some researches engineered flexible sensors using PVA ion-conductive hydrogels capable of monitoring various movements, including bending of the knee and fingers, even underwater [164]. With excellent ionic conductivity and high sensing stability, these sensors can also monitor electrocardiographic signals.

A highly sensitive hydrogel strain sensor capable of accurately detecting wrist flexion and transmitting electrical signals directly was developed. Improvements in sensing performance were achieved by incorporating tannic acid-coated cellulose nanocrystals (TA@CNCs) into conductive networks [165]. Placed on the radial artery, the developed sensor detects pulses with clear signals, advancing integrated electronic skin development. Self-powered hydrogels were applied to electromyography (EMG) [166], facilitating intuitive monitoring of human physiological health conditions through various methods. Lastly, a dual-network temperature-sensitive ion-conductive hydrogel was introduced for water-wearable strain sensors. By incorporating a PVP/TA/Fe3+-crosslinked network into a poly(N-isopropylacrylamide-acrylamide) (P(NIPAM-AM)) network [167], this hydrogel offers excellent stretchability and conductivity for sensor applications. The combination of wireless sensor network technology with wearable flexible sensors addresses the issue of constraints imposed by detection devices on traditional wearable flexible sensors during testing, further promoting the development of flexible wearable electronic devices. Lei et al. [168] introduced tannic acid-boric acid (TA-B) into a polyacrylamide/agar (PAM/Agar) double-network hydrogel matrix, forming a conductive hydrogel with antibacterial, transparent, and adhesive properties. The electronic skin sensor fixed on the fingers, neck, knees, or elbows can monitor and record changes in resistance caused by various bodily movements. When the hydrogel undergoes strain changes, the PAM/Agar/TA-B hydrogel electronic skin sensor synchronously changes with the bending and releasing of the moving part. In addition to the previously mentioned uses, there exist numerous other avenues worth exploring, such as the utilization of friction-based nanogenerators and supercapacitors. The allure of self-powered electronic gadgets lies in their pliability and ease of transport, particularly those hinging on adaptable electrodes. Supercapacitors represent a novel breed of energy retention apparatus, broadly categorized into pseudo capacitors (PC) and electric double layer capacitors (EDLC).

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Wang and colleagues [46] devised a triboelectric nanogenerator employing LiCl/PVA hydrogel (LP-TENG) (Figure 6e). When operated in a solitary polarity mode, the LP-TENG is capable of yielding an open-circuit voltage peaking at 276 V. Owing to its remarkable elasticity and superlative sensitivity, the LP-TENG finds favorable applications in motion surveillance (Figure 6f,g). Another team engineered a PVA-centric carbon supercapacitor [169], showcasing a steadfast voltage of 2 V, a capacitance measuring 24 F/g, and an energy density tallying 12.6 Wh/kg. Another team developed a self-repairing friction nanogenerator (HS-TENG) based on polyvinyl alcohol/agar hydrogel, demonstrating its ability to restore itself within approximately one minute [170]. The electric potential generated upon striking the HS-TENG serves the purpose of charging commercial LEDs, thus furnishing an uninterrupted power supply. Additionally, a renewable and malleable hydrogel electrolyte hinging on PAA and polyacrylamide (PAH) was divulged [171]. Under mild environmental conditions, the electrolyte stands primed for reshaping into any preferred configuration, with its original ionic conductivity enduring at 96% and 90% post the first and second reiterations, respectively. Employing polyacrylamide-chloride lithium (PAM-LiCl) hydrogel in conjunction with polydimethylsiloxane (PDMS) as constituent materials for electrodes and electrode strata [172], another team engineered a pliable transparent TENG (Figure 6h–j). This epidermis-esque TENG possesses a unilateral strain capacity scaling up to 1160 % and a translucency rating of 96.2%, effectively harnessing electrical energy at an immediate pinnacle power density of 35 $mW \cdot m^{-2}$, thereby converting kinetic energy stemming from human motion into electrical potential. In the realm of wearable applications, the resilience of self-repairing soft conductors underscores their categorical edge, manifested in their capacity to withstand distortions and rebound from injury.

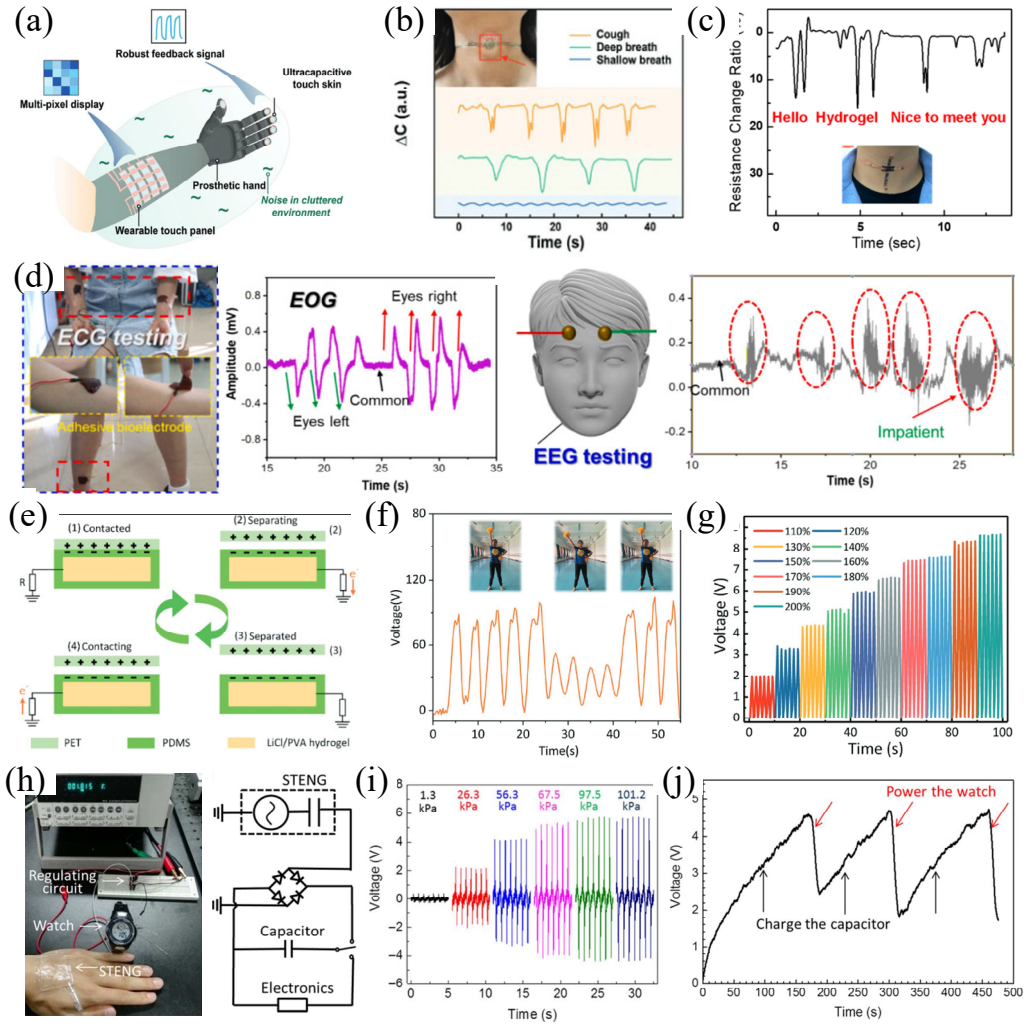


**Figure 6.** Several applications of conductive hydrogel in flexible wearable electronic: (a) Illustration for the functioning of the soft robotic skin system. (b) Electronic skin for real-time monitoring of breathing and coughing

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[161]. (c) The signal of the hydrogel sensor on the throat when saying "Hello, hydrogel, Nice to meet you" [91]. (d) Conductive hydrogel used for human ECG, EOG and EEG test signals [166]. (e) Schematic diagram of LP-TENG absorbing electrical energy. (f) and (g) Output voltage generated by LP-TENG self-powered sensors for body movement monitoring and output voltage at different stretching lengths [46]. (h) The physical diagram and equivalent circuit diagram of the electronic meter powered by the energy collected from STENG. (i) STENG serves as the voltage distribution under five different pressures. (j) The voltage distribution diagram of the capacitor that supplies power to the electronic watch by STENG [172].

## 5. Conclusion and prospects

In recent years, conductive hydrogels have demonstrated tremendous potential in tissue engineering and flexible electronics. As a multifunctional material, conductive hydrogels not only have the ability to mimic the structure of natural soft tissues but also possess excellent biocompatibility and biodegradability, facilitating cell adhesion and growth. In the field of tissue engineering, the application of conductive hydrogels is continuously expanding, especially in artificial tissue and organ regeneration. Its unique porous structure provides an ideal environment for cell colonization, while its conductivity enhances the microenvironment for cell culture, promoting tissue repair and regeneration. Furthermore, conductive hydrogels can serve as carriers for drug delivery systems, allowing for controlled drug release by modulating the gel's structure and chemical composition, thus offering new avenues and methods for tissue engineering. In the realm of flexible electronics, conductive hydrogels have garnered significant attention due to their excellent conductivity and flexibility. Flexible electronic devices often require contact with the human body surface, and conductive hydrogels possess properties that allow for excellent adherence to the skin. This makes conductive hydrogels an ideal material for smart wearable devices such as human motion sensors, sensory skins, and personal health monitors. By integrating conductive hydrogels with sensor technology, real-time monitoring and data collection of human physiological parameters can be achieved, offering new solutions for personal health management. Additionally, conductive hydrogels can be utilized for biological signal sensing and medical diagnostics, opening up new possibilities in the healthcare sector. In conclusion, conductive hydrogels hold vast prospects in the fields of tissue engineering and flexible electronics. With continuous technological advancements and innovations, it is believed that conductive hydrogels will bring more surprises and convenience to human health and life.

Despite the promising applications, conductive hydrogels still face several challenges in practical use. Firstly, while conductive hydrogels exhibit excellent conductivity and flexibility, their mechanical properties and stability need further improvement to meet the requirements of different application scenarios. Secondly, the biocompatibility and biodegradability of conductive hydrogels also require further research and optimization to ensure their safety and stability in the body. Moreover, simpler and more efficient methods for the preparation and processing of conductive hydrogels need to be developed to reduce production costs and increase production efficiency. Lastly, large-scale production and application of conductive hydrogels also face technical and market challenges, necessitating joint efforts from governments, industries, and academia.

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