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REVIEW

Conductive Polymer Based Hydrogels and Their Application in Wearable Sensors: A Review

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Hydrogels have been attracting increasing attention in wearable electronics, due to their intrinsic biomimetic features, highly tunable chemical-physical properties (mechanical, electrical, etc), and excellent biocompatibility. Among many proposed varieties, conductive polymer-based hydrogels (CPHs) have emerged as a promising candidate for future wearable sensor designs, with a capability of realizing desired features using different tuning strategies ranging from molecular design (with a low lengthscale of 10^{-10} m) to micro-structural configuration (up to a lengthscale of 10^{-2} m). However, considerable challenges remain to be overcome, such as the limited strain sensing range due to the mechanical strength, the signal loss/instability caused by swelling/deswelling, the significant hysteresis of sensing signal, the de-hydration induced malfunctions, the surface/interfacial failure during manufacturing/processing, etc. This review aims to offer a targeted scan of recent advancements in CPH based wearable sensor technology, from the establishment of dedicated structure-property relationships in the lab to the advanced manufacturing routes for potential scale-up production. The application of CPHs in wearable sensors is also explored, with suggested new research avenues and prospects for CPHs in the future also included.

1. Introduction

Personalized health monitoring system is considered as one of the pilot units for high-quality healthcare technologies since they can allow faster and effective diagnostics/treatment with a cost-effective manner.^[1-6] In this regard, health monitoring system based on wearable sensors (WSs) have attracted tremendous attention during the past decade, attributed to their facile/conformal interactions with the human body and long-term monitoring capabilities.^[7-13] WSs plays a critical role in the personalized healthcare sector as an effective transducer to convert the almost imperceptible changes (natural physiological fluctuations) into electrical signals such as impedance^[14-17], potential and current without affecting the user's motions.^[18-23] The key to realize the above functions is to develop a soft, biocompatible material that can work conformally on the human body and be compatible with its viscoelastic nature.^[24-26]

Hydrogels, a group of soft viscoelastic materials^[27-29], holds huge potentials to facilitate versatile structure-property relationships with their designable three-dimensional (3D) crosslinked network and widely available fabrication means ranging from molecular design^[30-32] (with a low lengthscale of 10^{-10} m) to micro-structural configuration (up to 10^{-2} m)^[33-36], which also profoundly affect the physical characteristics of hydrogels^[37, 38]

including the strength, toughness, ductility, corrosion resistance, electrical properties and so on. Another family of functional polymers is conductive polymers (CPs) (i.e. polyaniline (PANI), polypyrrole (PPy), and poly(3,4-ethylene dioxythiophene): poly(styrene sulfonate) (PEDOT:PSS)) represent a class of polymers that have a delocalized π system backbone enabling charge transfer along the polymeric chains, exhibiting electrical conductivity^[39-45].

CPs are kinds of organic materials with specific electrical and electrochemical performances, combining the advantages of carbon/metals as good conductors and polymers as low-cost and flexible materials. The unique combined properties of CPs have attracted large interests in various biomedical applications such as biosensors, neural probes, drug-delivery devices, bio-actuators and tissue-engineering scaffolds^[46]. PANI, PPy and PEDOT are some of the most widely used CPs, which have been considered as the alternatives to carbonaceous and metallic nanofillers for developing ECHs because of some of their special and unique properties such as high electrical conductivity, electrical properties, reversible doping/dedoping process, preparation simplicity, controllable chemical and electrochemical properties and environmental friendliness. CPs are a class of conjugated π polymers, possessing typical p-conjugated structures which endows electrons stay and transfer in their backbones^[47]. The conductive pathway is constructed by the freely moved delocalized π -electrons, thus providing an electrical route for mobile charge carries^[48], leading to a controllable conductivity^[49] (Table 1). Therefore, CPs are frequently utilized as an electrode for stimulation and recording, owing to the unique capability of chemical surface modification with physiologically active species to improve the biocompatibility and functionality. However, their application in WSs is limited because CPs are brittle and difficult to handle. Cooperate CPs with another polymer can

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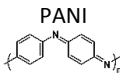
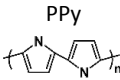
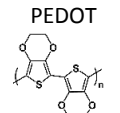
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REVIEW

combine the useful properties of both materials, in order to overcome the shortage of CPs while make full use of their advantages. Use appropriate method to construct CPHs can provide the increased solubility and better mechanical properties of CPHs, necessary for various Ws without significant compromise of their unique properties. Details are discussed in later sections.

Table 1. Properties of common CPs

Polymer	Properties	Limitations
 PANI	<ul style="list-style-type: none"> -Easy synthesis method -High stability -Low cost -Large specific surface area 	<ul style="list-style-type: none"> -Non-biodegradable, -Lack of flexibility -Limited solubility -Low processability
 PPy	<ul style="list-style-type: none"> -High electrical conductivity -Easy to synthesis -Stable in oxidized form -None toxic 	<ul style="list-style-type: none"> -Poor cycling stability -Brittle -Insoluble -Nontransparent
 PEDOT	<ul style="list-style-type: none"> -environmentally stable -electrochemically stable -Tunable conductivity -None toxic 	<ul style="list-style-type: none"> -Poor solubility -Limited flexible -Hard to process.

The conductive polymer-based hydrogels (CPHs) can be generated by employing CPs as scaffold for constructing continuous conductive networks in hydrogel, facilitating the transport and diffusion of electrons and charges.^[48-50] The resulted CPHs have been an effective building block for Ws, owing to their excellent conductivity (up to 10^3 mS m⁻¹), good biocompatibility, easy processing, environmental friendly property and cost-effectiveness.^[51-53] The CPs are the conductive unit of the bulk hydrogels, while the electrical conductivity in CPHs is entirely provided by their conductive networks formed with CPs. Enhance the content of CPs in CPHs could efficiently improve the bulk conductivity of CPHs, owing to that the isolated electronic transfer pathway gradually evolved into 3D conductive network, resulting to higher conductivity. Then the CPs contact to each other to construct a whole conductive network for electron transporting in relatively low-resistance. At this point, the conductivity of the bulk hydrogel depends on the inherent conductivity of the CPs. For example, Wang et al.^[54] demonstrated that the conductivity of optimized PANI based CPHs could achieve to 8.24 ± 0.57 S m⁻¹, which is close to that of pure PANI hydrogel^[55]. CPHs has also been utilized for flexible bioelectronics to improve the interface between biological and synthetic systems as well as the performance of medical devices^[56-65]. Compared to a blend of metallic/carbon particles that are island-type distributed in the polymer matrix, CPs chains are able to construct an ideal 3D interconnected conducting network. The interconnected conductive paths were suitable for accomplishing efficient tunnelling current, resulting in a high electro-conductivity.^[66, 67] However, there still remain technical obstacles: CPs have low water solubility, and hydrophobic CPs are easy to aggregate in hydrogel if there is no chemical bond between CPs and hydrophilic chains. As a result, (1) the continuous conductive

network in the CPHs is destroyed, significantly decreasing their conductivity, and (2) the aggregated CPs in the hydrogel tend to be stress concentration points when the CPHs are subjected to external force, decreasing their capacity to withstand significant stresses.

Considering these issues, continuous efforts have been devoted to improve the conductivity and mechanical performance of CPHs for high performance Ws, by building in a precise and robust the mechanical-electrical sensing response,^[68, 69] towards a wide range of applications.^[70-72] This review focuses on the construction of structure-property relationships of CPHs for the temperature or and mechanical sensing (**Figure 1**). We start by reviewing the development of novel network structure of CPHs, the associated strategies to realise these CPHs, and the various fabrication/integration approaches for CPHs-based Ws. Then, we discuss the state-of-the-art in CPHs-based Ws covering the sensing principles and the underlying mechanisms. A summary and outlook are then provided at the end with exploration of future design of CPHs-based Ws devices.

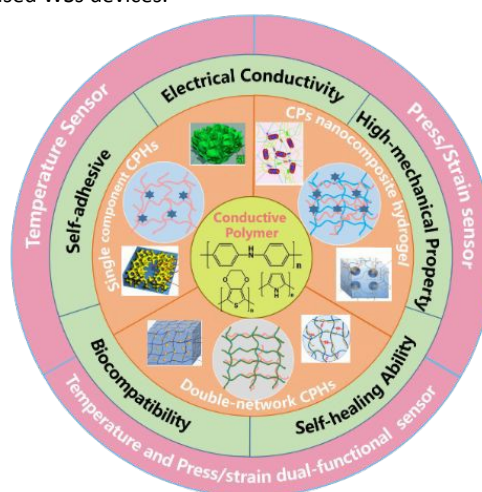


Figure 1. Schematic illustration of three categories of network structures of CPHs and their properties and applications being reviewed.

2. Fabrication of CPHs

CPs are hard to fabricate due to the aromatic structure and the intermolecular hydrogen bond between amine and imine groups, resulting in insolubility in most solvents.^[73-75] The insolubility leads to the inhomogeneous distribution of CPs in hydrogel networks, eventually compromises the mechanical and electrical properties. The homogeneous distribution of polymer chains to generate a continuous network is crucial to fabricate Ws with high sensitivity. According to the percolation threshold theory, uniformly distributed electronic CP chains in CPHs are contacting to each other when the concentration of the CPs arrived to their percolation threshold, building a continuous 3D conductive network. In this condition, the interior conductive network exceeded slightly under faint deformation, leading to sharply fluctuated conductivity. The increase in $\Delta R/R_0$ (For resistive-type strain sensors, $GF = (\Delta R/R_0)/\epsilon$, where $\Delta R/R_0$ is the relative resistance change, and ϵ is strain) is due to deformation and disconnection of the internal conductive network



within the film, corresponding to the high sensitivity^[76]. Otherwise, the island distributed CPs can't construct the continuous conductive network in CPHs, their $\Delta R/R_0$ are insensitivity to the external stimuli. Different synthetic approaches have been developed to address these concerns with processability by incorporating hydrophilic substituents and copolymerizing aniline with other monomers to generate soluble copolymers - this will be reviewed in the following section.

2.1 Synthetic processing of CPHs

The most effective way to synthesize CPHs network is the copolymerisation of CP monomers with non-conductive polymers. However, the copolymerized CPs are non-elastic with limited stimuli-responsiveness, due to the constrained mobility of chains caused by strong interactions between fillers and polymer matrix.^[50] To realize a continuous conducting network in CPHs, non-conductive polymer and CP precursors are combined and polymerized either in a two-step procedure (Figure 2a and 2b) or self-assembly in one step (Figure 2c) to merge CP into an insulating hydrophilic polymer matrix. The hydrophilic polymer acts either as a fragment of the main copolymer chains or cross-linking unit to bridge CPs.

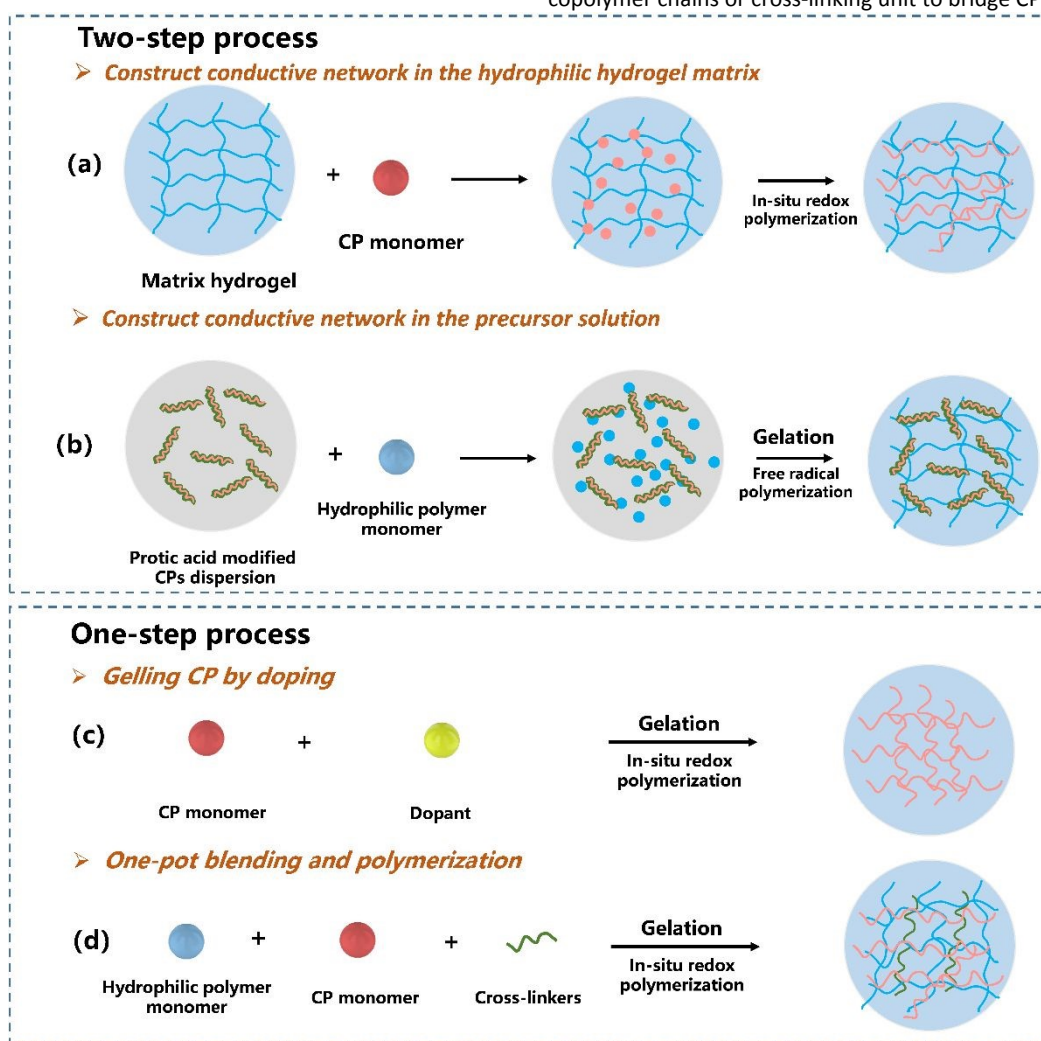


Figure 2. Schematic illustration of common synthetic approaches to prepare CPHs. (a) Construct conductive network in the hydrophilic hydrogel matrix. (b) Construct conductive network in the precursor solution. (c) One-step gelling CP by doping, and (d) one-pot blending and polymerization.

2.1.1 Two-step synthesis

The two-step synthesis of CPHs has two routes to fulfil a conductive network: (1) *Constructing conductive network in the hydrophilic hydrogel matrix* (Figure 2a). In this route, non-conductive hydrophilic polymer hydrogels are firstly formed by UV curing or free radical polymerization, such as poly(*n*-isopropyl acrylamide) (PNIAM), polyacrylic acid (PAA), and Polyacrylamide (PAAM or PAM)^[77-79]. Then, the CPs are introduced

into the hydrogel matrix to initialize the electron transfer pathways by absorbing the CP monomers into the hydrophilic hydrogel matrix, followed by the *in-situ* redox polymerization to create a conductive network of CPHs^[80-82]. (2) *Constructing conductive network then gelling* (Figure 2b). The CPs are treated with amphiphilic molecules and/or surfactants to enhance the water solubility and mixed with other monomers uniformly. Then, free radical gelation is performed to achieve CPHs.



REVIEW

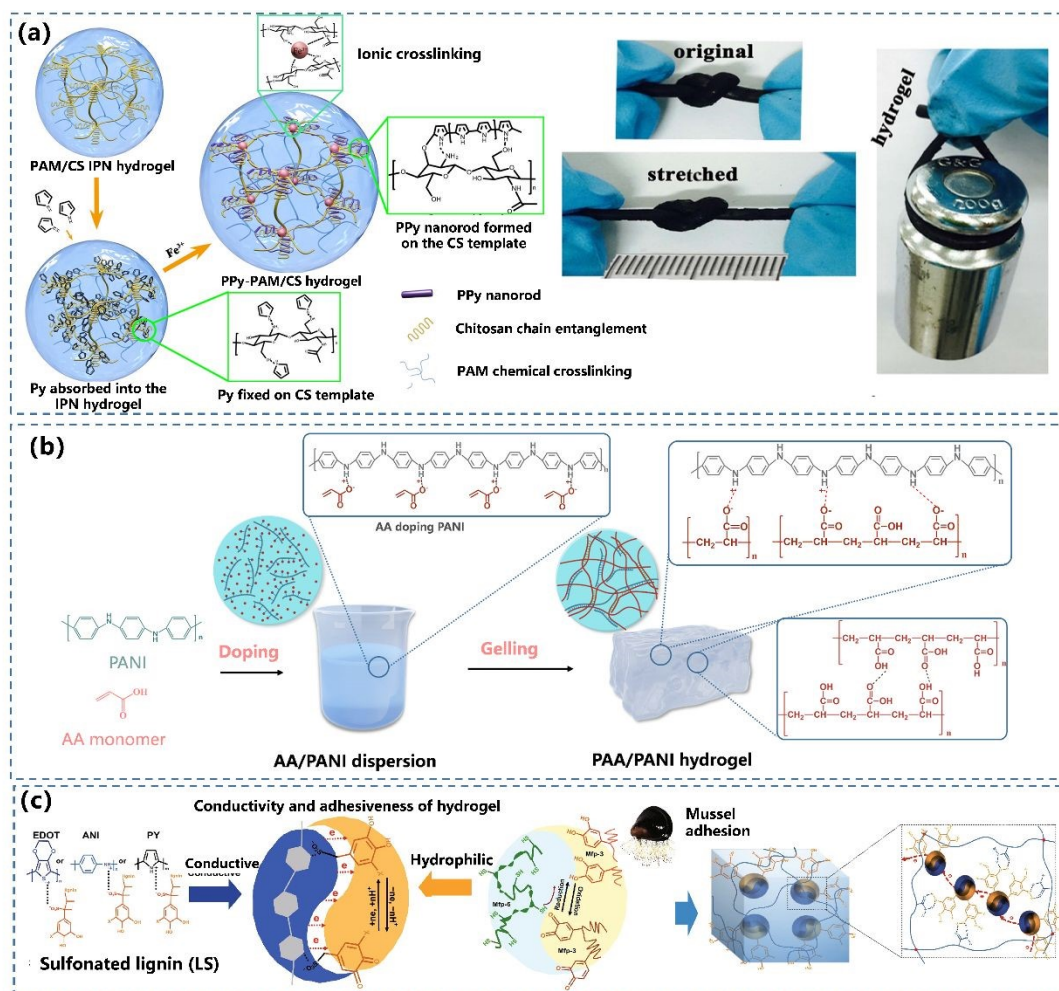


Figure 3. Construction of conductive network in the hydrophilic hydrogel matrix: The preparation and schematic illustration of structure for (a) PPy-PAM/CS hydrogel. Reprinted from ref. [83] with permission. Copyright 2018 American Chemical Society. (b) The process of "dope then gelling" strategy to prepare PAA/PANI gel. Reprinted from ref. [84] with permission. Copyright 2022 John Wiley and Sons. (c) Preparation of hydrophilic and redox-active CP/LS NPs hydrogels. Reprinted from ref. [85] with permission. Copyright 2020 Springer Nature.

Constructing conductive network in the hydrophilic hydrogel matrix

This is the most common route to build CPHs with a continuously conductive network. The hydrophilic hydrogel is soaked in a conductive monomer solution to allow monomer to sufficiently diffuse inside the porous structure of hydrogel and then the favoured network is generated by oxidative polymerizing of CPs^[86, 87]. Lu's group^[83] designed a tough and conductive PPy-PAM/chitosan (PPy-PAM/CS) hydrogel by employing CS framework as molecular templates to control the *in-situ* formation of PPy nanorods within the hydrogel networks (Figure 3a). The hydrophobic PPy nanorods were uniformly dispersed and merged with the hydrophilic polymer phase

in the PPy-PAM/CS hydrogel, resulting in outstanding conductivity, mechanical characteristics, and biocompatibility.

Constructing conductive network in the precursor solution

In this route, the morphology and physical/chemical properties of CPs can be manipulated using doping principles during the synthesis^[88, 89]. Our group developed a strand entangled supramolecular PANI/PAA hydrogel *via* a "dope then gelling" strategy^[84] (Figure 3b). The PANI nanorod was firstly doped with acrylic acid (AA) monomer, while the AA doped PANI possessed not only favourable dispersiveness but also enhanced conductivity. Lu *et al.*^[85] developed a universal technology to achieve CPHs by doping various CPs (PANI, PPy and PEDOT) with catechol/quinone contained sulfonated lignin, which significantly improved the water solubility of



CPs (Figure 3c). The obtained CPHs possessed high conductivity with a well-constructed electric path, and even a long-term adhesiveness. The down-sizing of CPs particle can also improve the dispersion of CPs into water. Yu's group^[90] cooperated PANI nanoparticles with poly(poly(ethylene glycol) methacrylate-co-AA) (P(PEG-co-AA)) network, to fabricate CPHs-based WSs with high stretchability, ultra-softness, excellent conductivity, and good self-healing ability. Benefiting from the homogeneous dispersion of nano-sized PANI and dynamically electrostatic interactions/hydrogen bonds between PANI and P(PEG-co-AA) scaffold, the obtained CPHs presented an excellent stretchability, a low modulus, and a fast self-healing performance. In addition, the subsequent demonstration of CPHs-based WSs show a unique strain detection, with a large range of linearity and outstanding anti-fatigue feature.

2.1.2 One-step process

Gelling CP by doping

One-step gelling CP by doping is conducted by directly cross-linking CPs chains without additional insulating hydrophilic polymer matrix (Figure 2c), feasible for constructing CPHs by introducing other cross-linkers with multi-functional groups such as PA^[55], copper phthalocyanine-3,4,4,4'-tetrasulfonic acid tetrasodium salt (CuPcTs)^[91] and amino trimethylene phosphonic acid^[92]. Owing to the six anionic phosphate groups, Bao^[55] have fabricated PANI based 3D porous CPHs by using PA as both dopant and gelator to interact with PANI chains. Moreover, plant-derived polyphenol tannic acid that contains multi phenolic hydroxyl groups can also act as dopant and crosslinker for conductive PPy hydrogel by constructing abundant intermolecular interactions^[93]. As a result, a soft, highly conductive, biocompatible CPH was developed.

One-pot blending and polymerization

CPHs can be fabricated by one-pot blending conducting precursors and hydrophilic precursors, followed by in-situ polymerization for gelation^[94-97] (Figure 2d). Among notable examples, Moussa *et al* demonstrated an universal method to achieve CPHs with a wide range of CPs including PANI, PPy and PEDOT^[98], while Zhao *et al*^[99] directly mixed PANI and crosslinked PVA to acquire stretchable CPHs *via* an ice-templated, low-temperature polymerization. The resulted CPHs performed an enhanced mechanical, electrical and electrochemical properties, with capability to monitor human motions at both large and small strains in real time. With a self-assembly process, the solution-processing issue is not a problem for CPs. PEDOT:PSS consisting of both positively charged conjugated PEDOT and negatively charged saturated PSS can be processed with this route^[100, 101]. Bao *et al* developed PEDOT:PSS hydrogels by using ionic liquid 4-(3-butyl-1-imidazolium)-1-butan-1-sulfonic acid triflate to increase the ionic strength of PEDOT:PSS solutions, yielding a remarkable enhancement in conductivity. Wu *et al*^[94] synthesized a highly stretchable supramolecular CPHs by doping poly(*N*-acryloyl glycinamide-co-2-acrylamide-2-methyl propane sulfonic) (PNAGA-PAMPS) with PEDOT:PSS. The PNAGA-PAMPS hydrogels displayed excellent mechanical properties such as a 1700% elongation at the breaking point, and a giant capacity due to the addition of PEDOT:PSS.

Notably, blending PEDOT:PSS with other non-conductive polymers always suffers from compromised mechanical and/or electrical properties. Zhao's team^[39] reported an interested pure PEDOT:PSS by interconnecting networks of PEDOT:PSS nanofibrils. Different from

the common used method, they used a simple method that mixed volatile organic solvent into aqueous PEDOT:PSS solutions followed by controlled dry-annealing and rehydration, constructing a high performance CPHs with controllable electrical conductivity, superior mechanical properties, and tunable isotropic/anisotropic swelling in wet physiological environments.

2.2 Manufacturing process of CPHs

Subject to the synthetic route of CPHs, it can also be manufactured through different processes such as casting/molding and 3D printing to achieve certain geometries. The manufacturing process can strongly influence the morphology of conductive networks and hence greatly affect their mechanical properties^[102, 103]. In the following section, we will review these two popular manufacturing processes.

2.2.1 Casting and molding

The casting and molding is a conventional approach to create CPHs with desirable morphology with advantages such as simplicity, high throughput and low cost.^[104-107] In this method, a hydrogel pre-solution is poured into a shaped mold, followed by polymerization which can be triggered by a range of stimulus, i.e. light, chemicals, or heat for gelling.^[108-110] This robust method has been widely used to fabricate simple and planar shaped two-dimensional (2D) or 3D structures for CPHs.^[111, 112] Zhou's group^[113] reported a hybrid PVA/PANI hydrogel with a fully physical cross-linked binary network acting as electrode of the pressure sensors. Based on the molding and casting process, hydrogels with reliefs were forged. The specific reliefs on the surface of 2D PVA/PANI effectively enhance the gauge factor of WSs, resulting to a high GF of 7.70 kPa⁻¹ and a sensing range of 0-7.4 kPa.

2.2.2 Additive manufacturing (3D printing)

3D printing is attractive as it can generate 3D structures in a layer-by-layer fashion through computer aided design (CAD)^[114, 115]. 3D printing technologies with appropriate "biocompatible inks" are potentially able to fabricate WSs with precise control. Extrusion-based and light curing-based 3D printing techniques have been demonstrated to print 3D structures by using DN hydrogels to achieve high stretchability, good toughness, and unique biocompatibility.^[116, 117] Different from the free-standing 3D CPHs structures that can be engineered by relocating the pre-printed designs on substrates through tiny nozzles, the 3D printing of CPHs can accomplish structures with complex and reproducible geometries with a resolution of a few hundred micrometers, which often relies on the shear thinning behaviour of the CPs or CP precursors. The applied pressure forces the hydrogel to flow, hence introducing shear forces, which allow material deposition in the desired patterns.^[118] A procedure consisting of a rapid cryogenization, lyophilization and redispersion has been developed by Yuk^[119], to achieve the printable PEDOT:PSS hydrogels with suitable rheological properties, thus to secure high resolution and high aspect ratio microstructures.

The 3D printing route can achieve electroactive CPHs with complex and defined geometry, by *in-situ* polymerizing CPs confined in a matrix framework. Yue *et al*^[120] mixed PAA, poly(ethylene oxide) (PEO) and aniline monomers to prepare a high-viscosity solution (Figure 4a) as an ink for 3D printing. The resulted CPH presented a large stretchability, high fatigue resistance and a high sensitivity



(7.10 kPa^{-1}). Besides of extruding the hydrogel precursors, light-based 3D printing has also been widely used to fabricate CPH. Jordan *et al.*^[117] demonstrated a stereolithography 3D printing strategy for constructing architected CPHs with complex lattice structures by manipulating the properties of precursor solution. The light-based 3D printed hydrogel precursor can also serve as the structural template for absorbing CP monomer. Fantino *et al.*^[121] coupled light-

based 3D printing with interfacial polymerization to get electroactive hydrogels with complex and defined geometry (Figure 4b) by using a digital light processing 3D printing system to fabricate a poly(ethylene glycol)diacrylate (PEGDA) 3D structure firstly, with subsequent polymerizing pyrrole monomer to create a conductive phase.

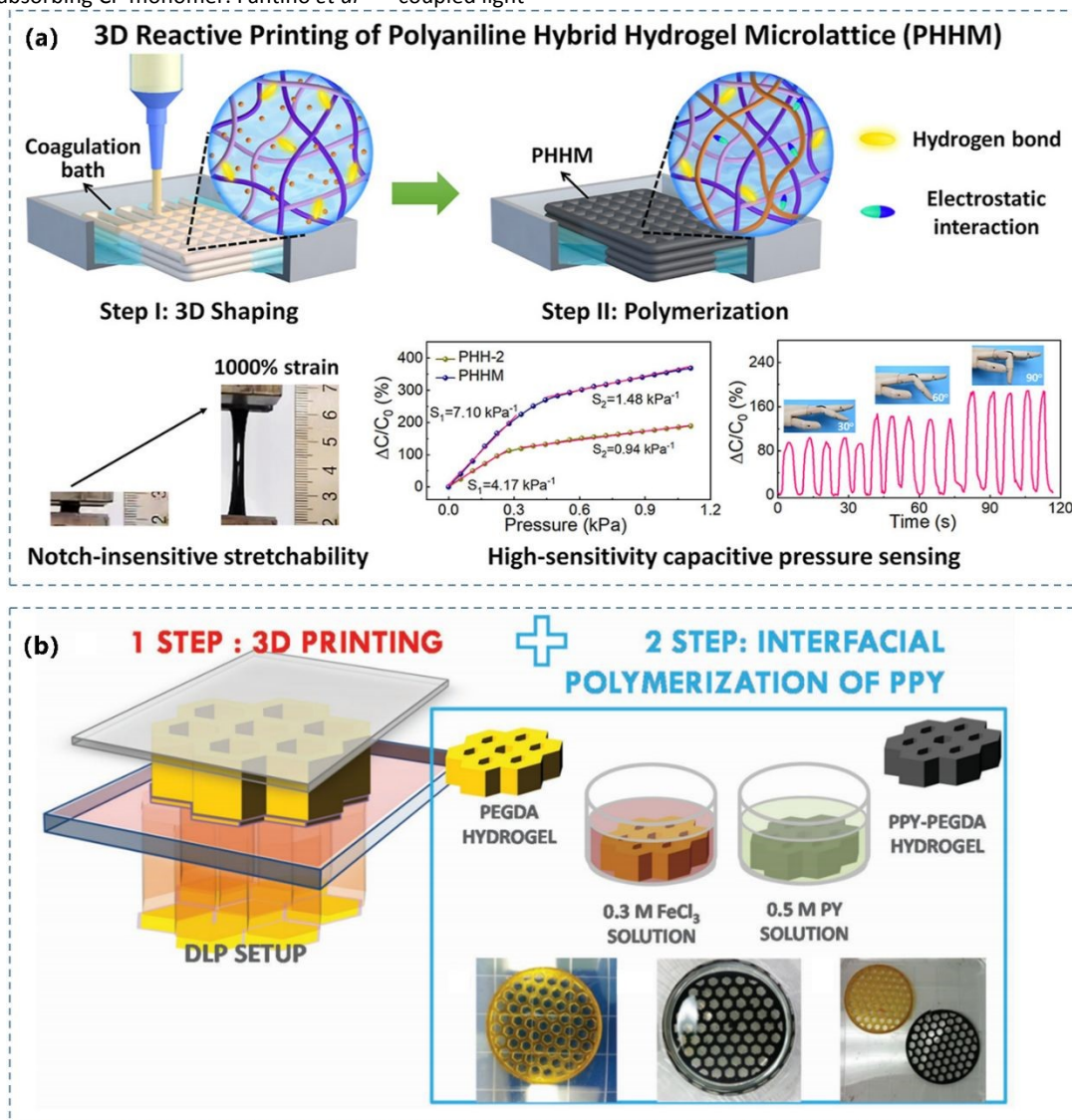


Figure 4. (a) Schematic of the preparation of 3D reactive printing of the intrinsically stretchable PANI hybrid hydrogel with pressure sensing performance. Reprinted from ref.[120] with permission. Copyright 2022 Elsevier. (b) Sketch of a light-based 3D printing based preparation process for the conductive hydrogel hybrids. Reprinted from ref. [121] with permission. Copyright 2018 John Wiley and Sons.

3 Network structure of CPHs

By far, many strategies have been established to deliver CPHs for WSs, which provide a broad perspective on designing and fabricating specific CPHs structures. We classify these network structures into three categories: (i) direct crosslinking the CPs and then constructing a single component CPHs,^[122] (ii) doping or blending the CPs into insulating polymer matrix, forming a conductive network with nanostructures embedded relatively uniformly. This structure is

called a CP nanocomposite hydrogel, which generally possesses better interactions between the hydrophilic polymer matrix and CPs, thus enhancing the mechanical properties.^[123, 124] (iii) double-network CPHs by composing the interconnected CPs and insulating polymer networks. The first network usually is a hydrophilic flexible matrix and hosts the polymerization of a conductive second



network^[86, 125], which is a crosslinked, modestly stretched network acting as an electronic transfer pathway^[126].

3.1 Single component CPHs

The delocalized π -electron system along the polymer backbone of CP leads to the inherent rigid macromolecular chains. The traditional method to construct single component CPHs is to introduce small molecules with multi-functional protonic acid groups^[127]. Recently, molecules with multiple functional groups have been reported to crosslink CP chains, resulting in an 3D network structured CP composite hydrogel. Superior to other dopants, phytic acid

molecules can link to multiple PANI chains, generating a strongly crosslinked structure (**Figure 5a**).^[92, 128] As described in section 2.1.2, Phytic acid (PA) is adopted as the gelator and dopant in the synthetic process of PANI, which possesses a unique structure of several anionic phosphate groups, to initiate a mesh-like hydrogel network. Bao and co-workers^[55] used phytic acid as PANI's cross-linker and dopant, to generate PANI hydrogel possessing a direct conductivity of 11 S m^{-1} (Figure 5b). Similar to PA, disc-shaped liquid crystal copper phthalocyanine-3, 4', 4'', 4'''-tetrasulfonic acid tetrasodium salt (CuPcTs)^[91] with tetra-functional groups was used to prepare a morphology-controlled nanostructured PPy hydrogel (Figure 5c).

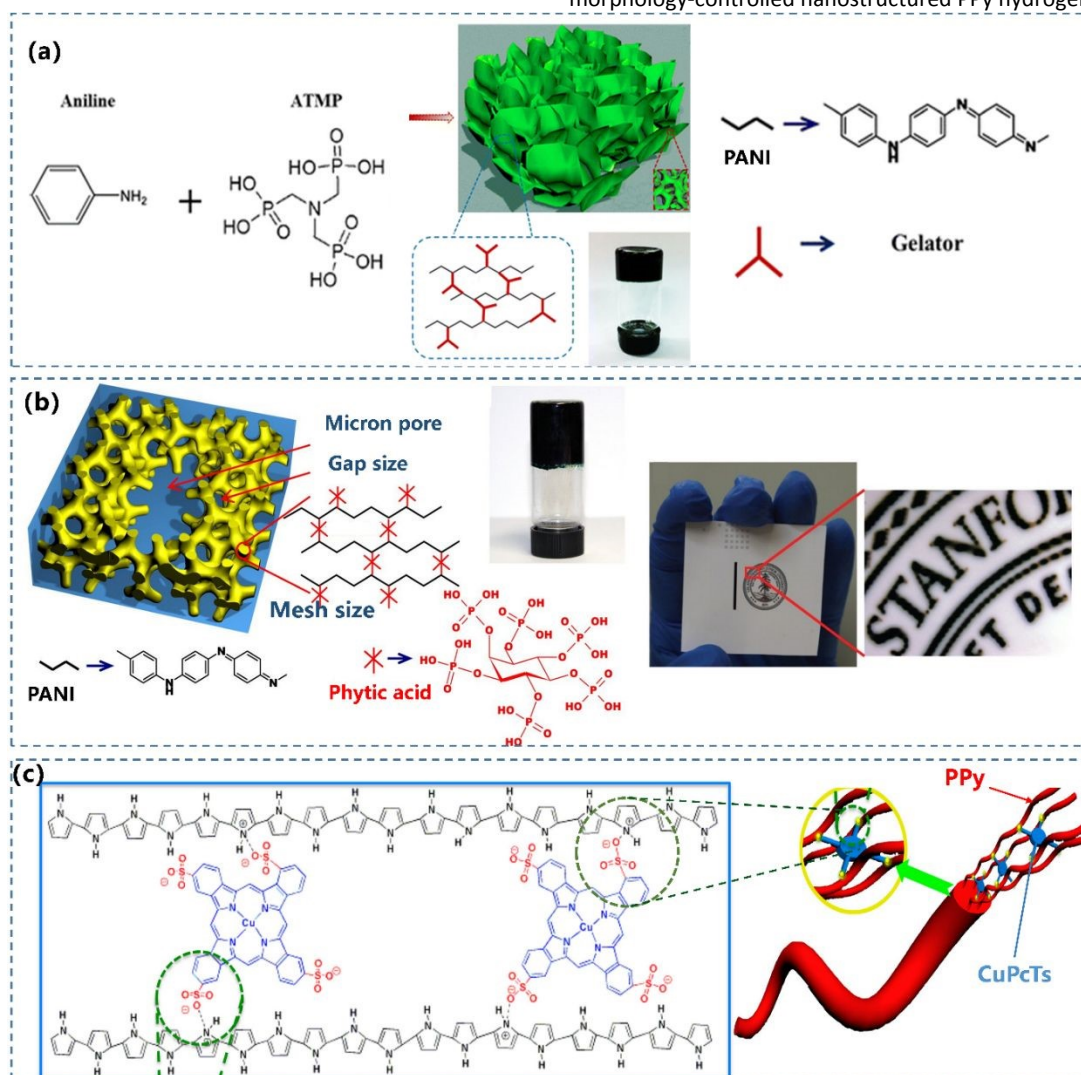


Figure 5. (a) Schematic illustration of the formation of 3D nanostructured PANI hydrogel and a photograph of the PANI hydrogel in a glass vial. Reprinted from ref. [92] with permission. Copyright 2016 Springer Nature. (b) Chemical structure and morphological characterization of phytic acid gelated and doped polyaniline hydrogel. The photograph of an ink-jet-printed image on a piece of glossy photo paper. Reprinted from ref. [55] with permission. Copyright 2012 National Academy of Sciences. (c) Illustration depicting the controlled synthesis of CuPcTs doped PPy hydrogel. Reprinted from ref. [91] with permission. Copyright 2015 American Chemical Society.

Besides of the cross-linking of inherent rigid polymeric chains of CP, elastic and conductive PPy hydrogels with mono-phase were synthesized by Zhang's group with an unique secondary growth mechanism^[129]. The formation of PPy occurred immediately upon the addition of oxidant to realize the incipient network via π - π interactions with low joint density. The insoluble PPy chains aggregate into spherical particles, coupling with the unreacted

pyrrole monomers formed protruded branches, resulting in a 3D building block. The optimized PPy hydrogel can be compressed by over 70%, and a fast full recovery within 30 seconds. This extremely elastic PPy hydrogel opens a window for developing elastic conducting hydrogels with applications in environmental engineering, biosensors, and regenerative medicine.

3.2 CPs nanocomposite hydrogel



Combining CPs with amphipathic molecules to increase the dispersibility of CPs is an efficient option to obtain high performance CPHs.^[130, 131] The CPH can be equipped with multiple characteristics, allowing both materials to retain their respective properties.^[132] There are two combinations of CPs and amphiphilic molecule: (1) To induce functional polymer with hydrophobic groups into hydrogel,

and associating the hydrophobic groups and CPs, thus dispersing the CPs uniformly in hydrogels, resulting in a continuous conductive network for CPHs (**Figure 6a**).^[85, 133] (2) To complex the CPs with hydrophilic molecules by non-covalent bonds to produce water-soluble CPs.^[134]

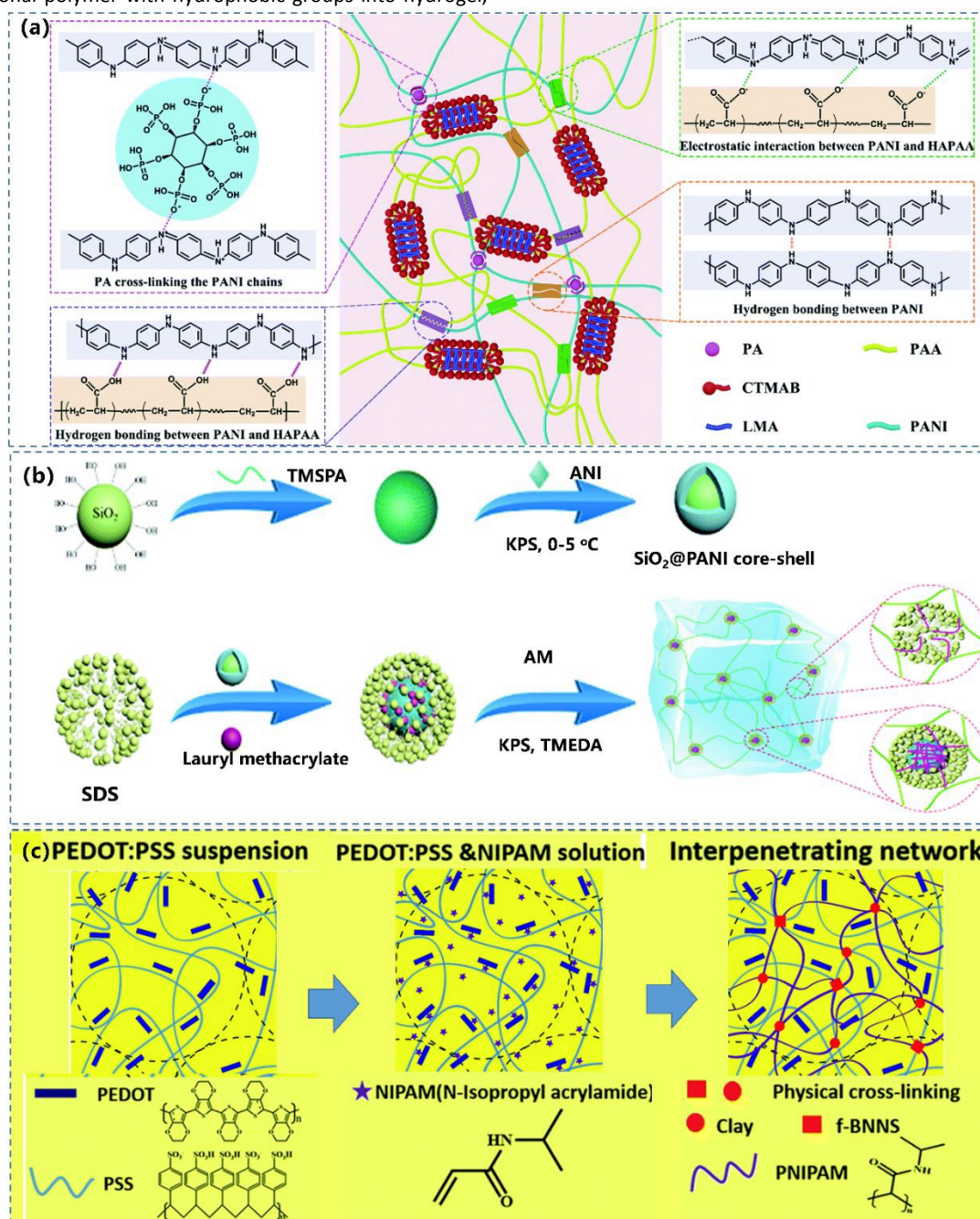


Figure 6. (a) Hydrophobic association of the physically cross-linked PAAN hydrogels and the interactions within the gel. Reprinted from ref. [133] with permission. Copyright 2021 Royal Society of Chemistry. (b) The preparation of SiO₂@PANI-P(AM/LMA) hydrogel. Reprinted from ref. [135] with permission. Copyright 2021 Royal Society of Chemistry. (c) Fabrication process of f-BNNS/PEDOT:PSS/PNIPAM hydrogels. Reprinted from ref. [136] with permission. Copyright 2019 Royal Society of Chemistry.

For the first strategy, the hydrophobic interaction in hydrogel prevents the CPs nanoparticles from aggregation. The modification of CPs with amphiphilic molecule to generate micro or nano-scale micelle can effectively support the dispersion of CPs into

homogeneous conductive component, to initialize the uniformly crosslinking to achieve continuous conductive network^[137]. Sun *et al.*^[135] designed a CPHs-based strain sensor by introducing PANI coated silica core-shell particles (SiO₂@PANI) into an acrylamide-



lauryl methacrylate copolymer (P(AM/LMA)) matrix (Figure 6b), where the hydrophobic interaction between the hydrophobic LMA in the P(AM/LMA) chains and the SiO₂@PANI core-shell particles induced stable and continuous conductive network. This resulted in a favourable electrical conductivity, high strain sensitivity with a Gauge Factor (GF) of 10.407 at 100–1100% strain, fast responsiveness (300 ms), and excellent durability (300 cycles).

Another approach is to produce soluble CPs by complexing them with hydrophilic molecules. The popularly used CPs nanocomposites hydrogel is PEDOT:PSS hydrogel^[136, 138, 139], with PEDOT domains placed in an electrically insulating PSS matrix under a loosely crosslinking association by hydrogen bonding. Zhao *et al.*^[139] reported a CP based hydrogel technique by interconnecting networks of PEDOT:PSS nanofibrils with excellent electrical, mechanical, and swelling properties (Figure 6c). The obtained pure PEDOT:PSS hydrogel fulfilled the desired properties for bioelectronic applications, including a high electrical conductivity, a low Young's modulus, and a tunable isotropic/anisotropic (de-)swelling in wet physiological environments. Apart from the hydrophilic polymers, hydrophilic micro- or nano- molecules are also imported to enhance the solubility of CPs. Lu's group^[85] doped catechol/quinone

contained sulfonated lignin to CPs to prepare a series of water-soluble CPs, including PANI, PPy and PEDOT. The resulted hydrogel presented a high conductivity with a uniformly distribution hydrophilic CPs and well-connected electric pathway. However, the CPs nanocomposite hydrogel is vulnerable to the phase separation between the CPs and hydrophobic polymer matrix, due to the intrinsic incompatibility.

3.3 Double-network CPHs

In the double-network CPHs, the system interlinks two polymer networks with adverse or immiscible properties to enable unique features such as high mechanical robustness, extreme stretchability, and bespoke chemical functionality.^[140–142] The double-network CPHs usually contains a densely cross-linked, fully stretched network that acts as a rigid skeleton, with the other network is cross-linked sparsely.^[143, 144] The interlinks between two network can be facilitated by covalent crosslinking and/or non-covalent bonds (i.e. hydrogen bond, hydrophobic or ionic interactions).^[145, 146] The dynamic cross-linking network enables the CPHs to form a reconfigurable structure with large deformation, which is critical for the mechanical properties, conductivity and sensing performance.

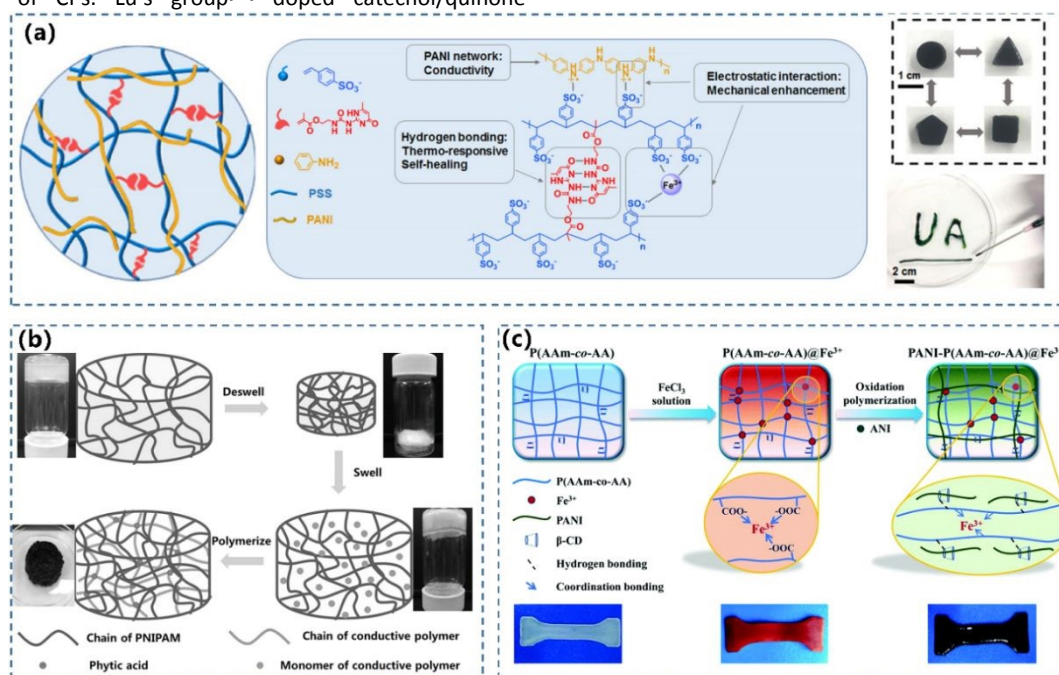


Figure 7. (a) The formation mechanism of supramolecular conductive PANI/PSS-UPy hydrogels, optical images of the hybrid hydrogels molded into different shapes and injected through a needle. Reprinted from ref. [78] with permission. Copyright 2019 American Chemical Society. (b) Synthesis of hybrid hydrogel composed of PNIPAM and conductive polymers. Reprinted from ref. [147] with permission. Copyright (2015) John Wiley and Sons. (c) Illustration of the fabrication of PANI-P(AAm-co-AA)@Fe³⁺ hydrogel. Reprinted from ref. [86] with permission. Copyright 2022 Royal Society of Chemistry.

The modulations of the topology for hydrophilic polymer and physically/non-covalent contacts between the networks can regulate the properties of CPHs-based WSs.^[54, 148] Chen's group used 2-ureido-4[1H]-pyrimidinone (UPy) to cross-link PANI in a hydrogen-bonded PSS network (Figure 7a)^[78]. Shi *et al.*^[147] synthesized PNIPAM/PANI and PNIPAM/PPy CPHs in the same process. Both the obtained CPHs were equipped with interpenetrating binary network structure and possessed a continuous transporting path for electrons. The PNIPAM/PANI and PNIPAM/PPy CPHs exhibited different electrical conductivity and thermoresponsive sensitivity,

owing to the different property between PANI and PPy (Figure 7b). Typical hydrophilic polymers including soft polymers (i.e. PVA^[51, 76, 77], PNIPAM^[147, 149] and PAm^[150, 151]), and rigid polymers such as crosslinked cellulose nanocrystals (CNCs)^[56, 152, 153] can be utilized to construct double-network CPHs. Ma *et al.*^[154] fabricated double-network CPHs by inducing supramolecular interactions between boronic acid groups on PANI and hydroxy groups on PVA. Because of the efficient energy dissipation, the CPH achieved good mechanical properties with a Young's modulus of 27.9 MPa, a tensile strength of 5.3 MPa, and an elongation at break of 250%. Tang' group^[86]



developed a synergistic dual network hydrogel PANI-P(AAm-co-AA)@Fe³⁺ composed of an iron-coordinated poly(AAm-co-AA) network and a conductive PANI network with adjustable mechanical properties and high sensitivity (Figure 7c). The host-guest interaction between β -cyclodextrin (β -CD) and PANI improves PANI's compatibility in a hydrogel substance, and leads to the formation of homogeneous interpenetrating networks.

4 Properties and applications of CPHs in wearable electronics

Significant progresses have been emerged in endowing CPHs with the desired characteristics in the application of WSSs, [87, 155-157] such as conductivity, mechanical properties, self-healing ability, self-adhesiveness, and biocompatibility. [158, 159] Recent advancements in this area is summarized in this section.

4.1 Properties of CPHs

4.1.1 Mechanical properties

The stress localization in soft electronics frequently happens during operation to cause the malfunction and system failure for the device, due to stretching, bending, tapping, or pressing [159-162]. Therefore, mechanical qualities are usually crucial to determine the performance of CPH-based WSSs, where numerous attempts have been made. [69, 72, 151] Considering the rigid nature of conductive

polymers, the constructive hydrogels are susceptible to long-term deformation, leading to fatigue and fracture after cyclic deformation [163]. The formation of an interpenetrating polymer hydrogel is believed to be a good strategy to endow the CPHs with long-term durability in mechanical properties, whereas the soft chain network can withhold the high elasticity of the interpenetrating network structure [164]. Sun and co-workers [165] proposed a dually synergetic network hydrogel composed of soft polymer PAAm and rigid conductive polymer PEDOT:PSS, the soft chains mechanically interlock with the rigid chains to provide the CPHs with ultra-wide sensing range of 0-2850%.

Upon the deformation, the dynamic noncovalent bonds (such as hydrogen bonds and the gelation network) normally behave as fragile, to serve as reversible "sacrificial bonds" to participate in energy dissipation via dissociation and/or reconstruction. Outstanding mechanical properties could be achieved via the mediation of inter- and intramolecular interactions based on the chemical and physical bonds. Dynamic interactions are prone to break to dissipate energy when CPHs is stretched, ensuring the hydrogels a fully and speedy recovery to their original states [166]. Xu et al [167] fabricated a polysaccharide-based conductive hydrogel through the fine-tuning at molecule-scale between conductive PEDOT and other polymers, which contributes to the significant improvement of mechanical behaviours.

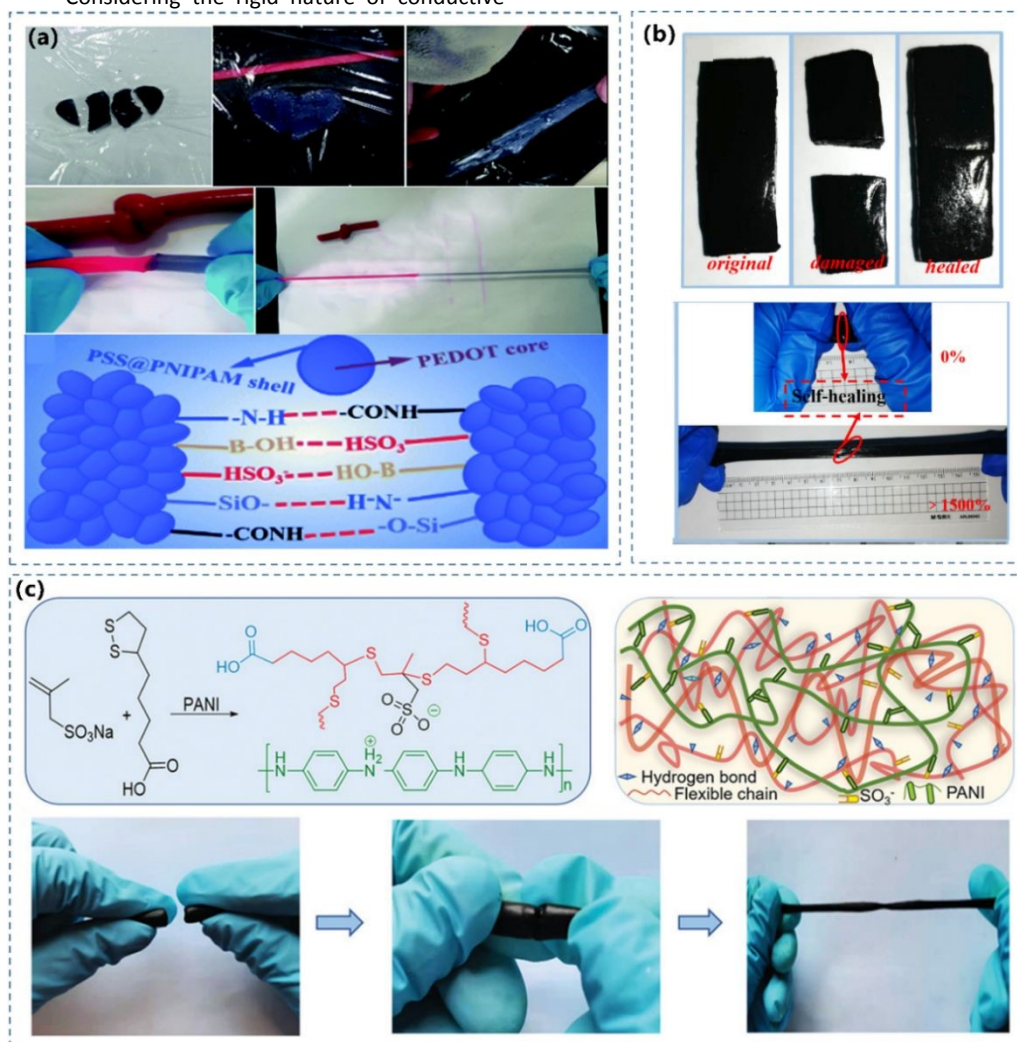


Figure 8. (a) The self-healing of obtained hydrogel and the illustration of mechanism^[136]. Reprinted from ref. [136] with permission. Copyright 2019 Royal Society of Chemistry. (b) Self-healing test of CNC-PANI/PVA hydrogels with healing time of 30 s. Reprinted from ref. [153] with permission. Copyright 2020 Elsevier. (c) The diagrammatic illustration of structure and characterization of the CPHs. Reprinted from ref. [168] with permission. Copyright 2021 John Wiley and Sons.

4.1.2 Self-healing

Self-healing is an appealing feature of CPHs to allow a quick restoration of mechanical and electrical characteristics, an extended lifespan of CPH sensors and a dramatical reduction on the cost (Figure 8a and 8b)^[136, 169-172]. The self-healing mechanisms can be classified into two types, intrinsic and extrinsic. The addition and polymerization of healing agents, such as some natural polymers like gelatin, carrageenan, and agar, is the fundamental extrinsic self-healing mechanism^[173, 174]. The intrinsic self-healing hydrogels are usually double-network hydrogels can undergo reversible fracture and reconstruction. The main driving forces are the dynamic non-covalent bonds (i.e., hydrogen bonds, electrostatic bonds, hydrophobic interactions, π - π interactions, and host-guest interactions) and supramolecular interaction.

Incorporating CPs into supramolecular network is effective to bring self-healing feature to hydrogel, because multiple dynamic interactions can be established between the conductive polymer and the other polymer network. Based on the good hydrogen donor of PANI, a self-healing PANI-based hydrogel was fabricated by introducing hydrogen acceptor PAAm.^[175] The fractured PANI hydrogel can be restored to its original structure ascribed to the reconnection of hydrogen bonds. Li *et al.*^[168] prepared double network CPHs through integrating rigid PANI chains with flexible sodium methallyl sulfonate functionalized poly (thioctic acid) polymer chains (Figure 8c). In addition, reversible covalent interactions are also intriguing for imparting hydrogel with self-healing capability. Wang *et al.*^[176] prepared a self-healing CPHs with long lifespan by rearranging the Diels-Alder covalent bonds between polymer chains.

Integrating self-healing capability and mechanical toughness together in a single structure is intriguing due to the conflicting demands. Strong molecular interactions are indispensable for imparting hydrogels with robust toughness, whereas high cross-linking density of networks are detrimental to the mobility of polymer chains, restricting the possibility of self-healing of hydrogels. As such, most self-healing CPHs are fabricated based on the incorporation of multiple weak molecular interactions, which will sacrifice the mechanical strength and toughness of CPHs. In light of this trade-off effect, a self-healing CPHs that balance self-healing and the mechanical properties (toughness and stretchability) were delivered by Su *et al.*^[133] They constructed dynamic interfacial interactions (including hydrogen bonding and electrostatic interactions) between the conductive PANI and hydrophobic association poly(acrylic acid) (HAPAA) network to elaborate interfacial interactions without sacrificing their self-healing capability. On one hand, the dynamic interfacial interactions constructed multiple energy dissipation mechanisms for endowing the CPHs with exceptional mechanical properties. On the other hand, the multiple reversible dynamic interactions between two networks can reassociate quickly after break, therefore endowing HAPAA hydrogel with good self-healing ability.

4.1.3 Self-adhesive

WSs with self-adhesive property ensure a reliable and conformal contact with human skin, reduce the interface resistance and stabilize the stimuli detection^[177-181]. To equip the CPHs with self-adhesiveness, the introduction of some moieties that can adhere to different substrates is of particular importance. The incorporating catechol-based molecules for providing hydrogel with adhesiveness that inspired by muscle has gained great attentions.^[163, 182, 183] Lu *et al.*^[89] integrated catechol molecules to dopamine, to synthesis PPY-based CPHs. Catechol moieties imparted the hydrogel with excellent adhesiveness, and high conductivity due to the effective paving of conductive pathways based on the dopamine-decorated PPY nanofibrils. Meanwhile, through tuning the covalent and noncovalent content, dynamic network based on PEDOT:PSS can be well controlled. As such, the resultant CPHs exhibits durable adhesion with high electroconductivity and cytocompatibility^[167].

The hydrated environments (e.g., sweating, raining and swimming) endangers the performance of WSs by deteriorating the interfacial adhesion^[184]. Incorporating hydrophobic moieties into hydrophilic polymer network is an effective way to tackle this challenge^[185]. Gao and co-workers^[186] designed a hydrogel with robust underwater adhesion and reliable fatigue-resistance capability, by copolymerizing hydrophilic monomers and hydrophobic monomers. The formation of hydrophobic aggregation expels the water and guarantee the direct contact of adhesion groups attached to hydrogel with substrates. In addition, there is another approach to introduce a several nanometer-thick hydrophilic adhesive layer onto the CPHs via interpenetrating polymer network, to provide hydrogel with strong underwater adhesion without sacrificing their mechanical and electrical characteristics^[187].

4.1.4 Electrical conductivity

A high electrical conductivity is always required for electronic readout purpose. CPHs are made up of CPs with intrinsic conductivity and soft hydrophilic polymer networks. The insulating hydrophilic polymer networks serve as scaffolds, while the CPs provide the conductive pathway. The electrical conductivity significantly affects the electrical transmission and sensitivity of CPHs. Chen's group generated an interpenetrating PANI/PSS network, with a good conductivity, stretchability, and self-healing ability by optimizing the concentration of PANI^[78]. Fu's group^[54] demonstrated that the electron transit in hydrogels is dependent on the tunnelling effect when the induced CPs are at low concentration. Continuous conductive networks can be established by further improving the concentration of PANI to reach their percolation threshold, the PANI chains are contact with each other and thus creating electron transit channels with low resistance, leading to a high conductivity of 8.24 S/m.

The electronic conductivity originates from the unique conjugated π -structure of CPs, which are organic conductors that depend on electronic conductivity. There are two main methods to improve the conductivity of CPHs: On one hand, enhance the content of CPs in CPHs could efficiently improve their conductivity, owing to that the



isolated electronic transfer pathway gradually evolved into 3D conductive network, which proved higher charge transport speed, leading to higher conductivity. For example, both Wang^[80] and Jin^[151] demonstrated that improved conductivity of CPHs can be obtained by enhancing the feeding ratio of CPs monomer, and the conductivities of the optimized samples are tens of times than that of the virgin hydrophilic hydrogels. The whole conductive networks are further created when the concentration of CPs is improved to their percolation threshold, forming relatively low-resistance paths for electron transportation due to the contact of conductive materials^[188]. Notably, the conductivity of CPHs will approach to a plateau value once the content of CPs exceeds the percolation threshold. On the other hand, Doping is a common process to improve the CPHs' electrical properties^[88]. Doping CPs with proton acid creates bipoles and dipoles along the polymer chain, transferring more carriers in the form of electrons or holes, which significantly improves the conductivity of conducting polymers. Milakin et al. demonstrated that higher content of dopant leads to increased conductivity^[128], this can be attributed to higher resistance to deprotonation of a phytic acid doped PANI which occurs during washing of the resulting cryogels with water. Moreover, the conductivity of CPHs is highly dependent on the type and concentration of dopants. Wang et al.^[91] synthesized PPy hydrogels with three different types of dopants and measured the conductivity of the obtained PPy hydrogels as 7.8, 0.4, and 0.06 S cm⁻¹, respectively.

4.1.5 Biocompatibility

The WSs can comprise various types of physical sensors, in detecting a change in a physical stimulus and then generating a signal to be measured or recorded. To accurately acquire human movement, it is essential to adhere the CHPs directly to human skin. Considering the direct contact between CHPs and skin, endowing the CHPs with good biocompatibility is of critical importance^[189]. CHPs have been developed as wound dressing^[89, 190], tissue engineering^[191] and WSs^[153, 192] due to their good biocompatibility and easy synthesis.

4.2 CPHs for wearable sensors

In general, the ideal conductive hydrogel for sensors should be stretchable, tough, conductive, self-healable, and highly sensitive to external deformations. CPs chains are able to construct an ideal 3D interconnected conducting network for providing an all-organic, continuous conducting path compared to that of noncontinuous,

point-to-point connected conducting network derived from inorganic nanoparticles^[75], could be helpful for fabricating press/strain sensor with high sensitivity and linearity, which are pivotal for the feasibility and accuracy of hydrogel sensors^[54]. As compared with other conductive fillers-based hydrogels, they usually show a nonlinear dependence of sensitivity on strain, presumably due to the damage of noncontinuous conductive network during stretching^[193]. Moreover, organic and polymeric nature of CPs enables diverse chemical modification and cross-linking strategies, resulting to extraordinary flexibility and self-healing ability^[22, 133, 194]. On the contrary, high stiff fillers such as metallic/carbon materials would decrease the self-healing ability of the hydrogel by restricting the mobility of polymer chains, the ability to restore equivalent sensing functions after healing from severe damage is also limited. Island-type distributed inorganic fillers in the hydrophilic polymer matrix are also challenging to balance the trade-offs among mechanical, electronic, and self-healing properties. A detailed list of examples of CPH-based sensors can be found in Table 1.

A range of CPHs based WSs have been developed that can respond to pressure, temperature, and metabolites, among other stimuli.^[188, 195-198] Various advanced wearable sensors possess the ability to detecting at least two stimuli at the same time. In this section, we review recent developed sensing modes in CPHs based WSs, such as resistive, capacitive, and triboelectric typed sensing modes. Moreover, the recent advances in wearable sensor applications including thermal, press/strain sensors, temperature and pressure/strain dual-functional sensors, gas sensors and humidity sensors will be reviewed.

4.2.1 Sensing mechanism

CPHs-based WSs can adaptively respond to the human motion^[199]. The interconnected conductive network and/or interactions between the CP chains and hydrophilic polymer matrix in CPHs enhance the WSs' responsiveness. Two representative categories are resistive sensor and capacitive sensor.

Resistive sensor

Resistive sensing mode is one of the most widely used sensing mode in WSs owing to its relatively high sensitivity^[200-202] (Figure 9a). This type of WSs, usually made by in hydrogels with low concentration of CPs^[203], depends on the change of configuration in conductive networks, and bring high sensitivity *via* contact-resistance effect and tunnelling effect. Resistive sensor can be divided into two categories: (1) piezoresistance sensor, typical for sensing press/strain; (2) thermoresistive sensor for sensing temperature.

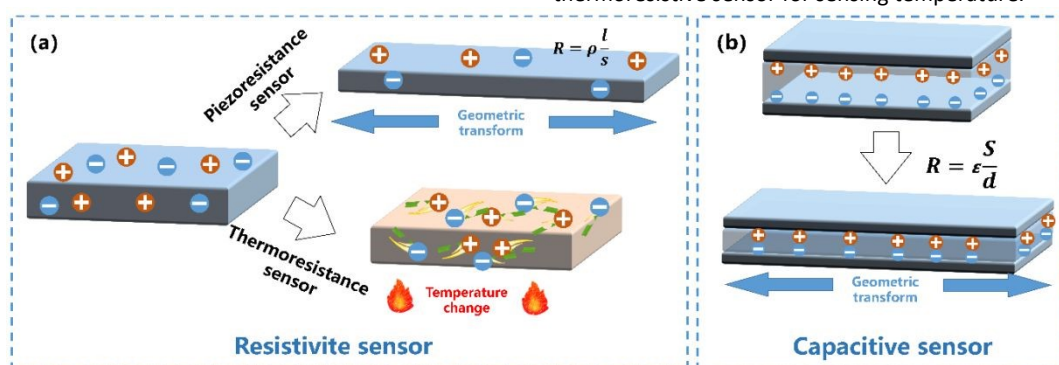


Figure 9. Schematic illustration on the structure and working mechanism of (a) Resistive sensor, and (b) Capacitive sensor.



Table 1. A detailed list of examples of CPH-based sensors

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Application	CPHs	Conductivity	Dopants	Stretchability	Specific device metrics	Ref.
Press/strain sensor	PAM/gelatin/PEDOT: PSS	-	PSS	2850%	GF=0.46-1.58	[165]
	poly(HEAA-co-SBAA)/PEDOT:PSS	0.625 S/m	PSS	4000%-5000%	GF=2	[195]
	PANI/PVA/PA/GA	3.4 S/m	PA	670%	GF=3.4	[77]
	PPy/PU	0.055 S/m	-	592%	-	[204]
	PEDOT: PSS-PVA	-	PSS	~370%	GF=4.07	[32]
	PANI/PAA/glycerol	-	PAA	~1500%	TCR=-0.0164 °C ⁻¹	[22]
	PNIPAM/CMCS/MWCNT/PANI	5.5 S/m	HCl	~400%	-	[205]
Humidity sensor	HAPAA/PANI/Gly	~3 S/m	PAA	2378%	TCR=-1.28% °C ⁻¹	[206]
	PNIPAM/PPy	0.8 S/m	PA	-	-	[147]
	PEDOT:PSS	-	PSS	-	RH range:2-90%	[207]
	CCNF/PEDOT:PSS	58 mS/m	PSS	929%	RH range:0-85%	[208]
Gas sensor	CeO ₂ @PANI	-	PA	-	6.5-50 ppm (NH ₃)	[209]
	PEDOT:PSS/IrO _x	-	PSS	-	17-7899 ppm (NH ₃)	[210]
	PEDOT:PSS	-	PSS	-	13-21 Vol% (O ₂)	[211]
Energy storage system	PANI	0.11 S/cm	PANI	-	450 F g ⁻¹ (0.5 A g ⁻¹)	[55]
	PANI/PA/PVA	91.0 mS/cm	PA	180%	2097 mF cm ⁻² (2 mA cm ⁻¹)	[99]
	PPy-CPHs	2.3 S/cm	p-TSA	-	560 F g ⁻¹ (0.75 A g ⁻¹)	[212]
	PEDOT-PVA	0.9 S/cm	boric acid	>400%	75.9 F g ⁻¹ (0.29 A g ⁻¹)	[213]
	PEDOT:PSS/silk fibroin	1 mS/cm	PSS	-	1.1 F cm ⁻² at 0.5 mA cm ⁻¹	[214]

The mechanical deformation on piezoresistance sensors leads to the change in resistance^[125, 215]. Under a small deformation, the tunnelling distance of the conductive network in piezoresistance sensor exhibits minute variation. Subsequently, some tunnelling channels increase linearly in tunnelling distance with increase in the applied force. A stronger signal is produced when the tunnelling distance exceeds the cut-off distance. Some direct connections between the conducting materials are broken^[216, 217]. Thermoresistive typed sensor can rapidly respond to the thermal stimuli by changing its own resistance. The sensing mechanism of thermosensitive CPHs can be classified into two groups: (1) CPH-based WSs to detect the body temperature with reversible temperature response^[218-220]. The mechanism is fulfilled by cooperating CP with thermal-sensitive polymer such as PNIPAM^[149], to switch the conductive network's tunnelling distance by harnessing the thermal-sensitive polymers' capacity. (2) CPs inherently have

excellent thermal expansion^[22, 221], to rapidly react to instantaneous temperature fluctuation of human's body. Temperature rise quickly accelerates carriers hopping between inter-fiber junctions, resulting in an ultrafast response in signal^[222].

Capacitive sensor

Compared to the resistive sensor, capacitive sensor possesses higher linearity, enhanced hysteresis, and faster response, for sensing press/strain^[223]. A capacitive sensor can be constructed by sandwiching a dielectric layer between two conformable conductive layer^[113, 120] (Figure 9b). The formula $C = \epsilon S/d$ can be used to determine the capacitance (C) of capacitive sensor, where the ϵ , S , and d are corresponding to the permittivity, contact area, and thickness of the dielectric layer, respectively. The sensors will respond the external stimuli by adjusting the interval between two parallel plates, resulting to an output capacitance signal change^[224]. For example, Mo and coworkers^[225] capacitive-type strain sensor



based on alginate/PAAm hydrogel and conductors and a semi-interpenetrating network structural organogel dielectric. The obtained capacitive strain sensor possessed extremely operational stability and the fast response speed for detecting small physiological signals and large-range human motions real-timely. However, for hydrogel-based press/strain sensor, most of them in literature belong to resistance sensors while minorities are capacitive sensors^[30]. Notably, there's little CPHs based capacitive press/strain

sensor as of today, we think this is mostly due to that the sensing mechanism of the capacitive sensor is oriented from the electronic double layer (EDL) forming at the electrolyte-electrode interface^[226], while the faraday pseudocapacitive behavior of the CPs will disrupt the linear sensing signal of the EDL capacitive sensor.

4.2.2 Sensing applications

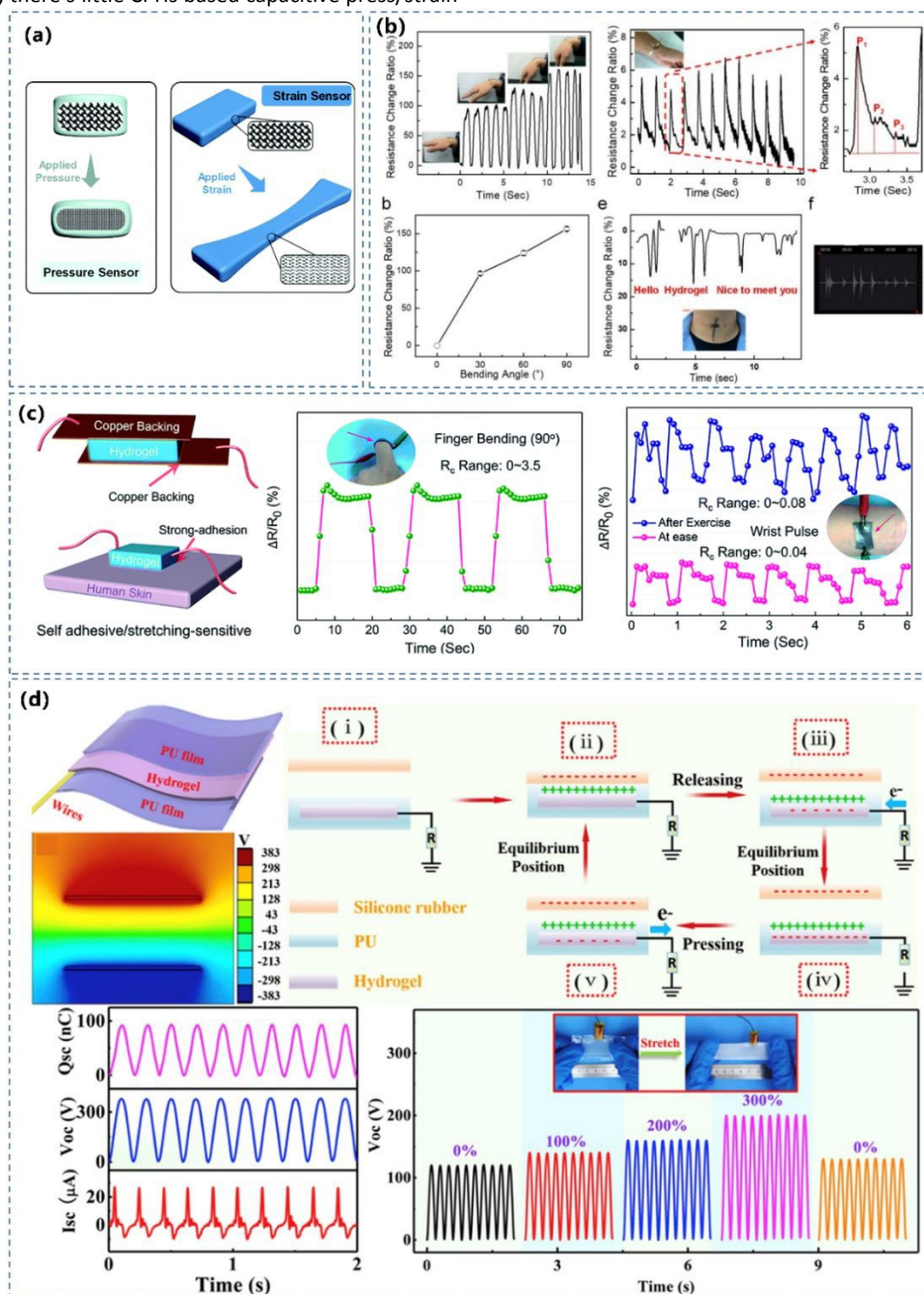


Figure 10. (a) Schematic illustration of pressure and strain sensors. Reprinted from ref. [125] with permission. Copyright 2020 Royal Society of Chemistry. (b) Signals from CPHs sensors on the wrist, the wrist pulse and on the throat of a volunteer speaking "Hello", "Hydrogel" and "Nice to meet you". Reprinted from ref. [54] with permission. Copyright 2018 American Chemical Society. (c) Schematic illustration of stretching-induced and pressure-induced hydrogel strain sensors and sensing performance. Reprinted from ref. [195] with permission. Copyright 2020 Royal Society of Chemistry. (d) CPHs-based highly stretchable (300% strain) TENG device and its energy harvesting performance with giving a short circuit current (I_{sc}) of 26.9 μ A, open circuit voltage (V_{oc}) of 383.8 V and short-circuit transferred charge (Q_{sc}) of 92 nC. Reprinted from ref. [165] with permission. Copyright 2020 Elsevier.



REVIEW

Pressure/strain sensors

Pressure/strain sensors are popular which generate an electrical signal dependent on either pressure or strain (**Figure 10a**). The CPHs-based press/strain sensors are normally made by interpenetrating conductive polymer-based hydrogel networks by cross-linking various non-covalent bonds^[112, 227-229]. Sensing sensitivity, response speed, and long-term durability under force loading are key merits for pressure/strain sensors, where a rapid response and low hysteresis are particularly needed^[125, 230].

The piezoresistive sensors are 'starring' players in pressure/strain CPH sensors, due to the facile fabrication, simple read-out mechanism and superior sensitivity^[54, 231]. The piezoresistive effect is a mainstream mechanism in the pressure/strain sensor, and a higher value of GF can endow a better precision of the sensation. Zhang's team^[195] synthesized a conductive hydrogel by interpenetrating conductive PEDOT:PSS polymers and zwitterionic poly(HEAA-co-SBAA) polymers (**Figure 10c**), capable of change its conductivity under deformation. Xu group^[232] prepared PANI based hydrogel with potentially allows for large-area, low-cost fabrication WSSs. The prepared sensor had a sensitivity of 37.6 kPa⁻¹ in a range of 0-0.8 kPa and presented ultrahigh sensitivity even under high load (1.9 kPa⁻¹ at 5 kPa). Bao's group^[233] developed an ultra-sensitive pressure sensor based on an elastic, microstructural conducting polymer thin film

Temperature sensors

The temperature of human body has been a primary parameter to indicate the status of fitness of individual^[237]. Precise, continuous and real-time monitoring of temperature for human body is critical to reveal the healthy state, to prevent the evidences for early diagnosis of diseases such as cardiovascular diseases, pulmonological diagnostics and infectious diseases such as COVID-19^[238]. Among those developed CPHs^[239-241], CPHs for detecting of temperature through resistive changes have attracted considerable attentions. The conventional fabrication of CPHs temperature sensors is to cooperate conductive polymer with thermal sensitive polymer. The CPHs' bulk electrical resistivity responds to absolute temperature, with the slope of curve relating to sensitivity or the temperature coefficient of resistance. Yu et al^[147] synthesized thermally responsive and conductive hydrogels by cross-linking PANI with phytic acid in PNIPAM matrix. The resulted PANI/PNIPAM hybrid hydrogels demonstrated a unique combination of excellent electrical conductivity, high thermo-responsive sensitivity, and increased mechanical capabilities.

Temperature and pressure/strain dual-functional sensors

The transfer speed of carriers between inter-molecular junctions can quickly increase as the temperature rises. The conductivity and temperature coefficient of resistance were improved since the thermal vibration triggered electrons could produce tunnelling current through nearby PANI junctions^[149, 193]. Ge et al.^[22] reported a

made from a PPy hydrogel. The micro-patterned PPy-based hydrogel pressure sensor has an ultrahigh sensitivity in the low-pressure regime and can detect pressures as low as 1 Pa.

Recently, triboelectric nanogenerators (TENG) lead an energetic trending development of WSSs by, not only efficiently harvesting the everyday body motion and powering various portable electronics, but also detecting pressure/strain stimulus in the process of generating electricity. The hydrogel-based TENG sensors generate potential signals without any external power supply with a number of superiorities such as, lightweight, prominent mechanical stability and facile cost-effective device structure. Shen and co-workers^[165] used PEDOT:PSS as conducting component, combined the rigid physically cross-linked gelatin, tough chemically cross-linked PAAm to construct stretchable and conductive hydrogels (**Figure 10d**). The device was not only served as a stretchable TENG for effective energy harvesting and self-powered wearable device, but also exhibited favorable pressure/strain sensing ability, including high sensitivity (GF=1.58), rapid responsiveness (200 ms), broad sensing range of 0-2850%, and stable/reproducible output sensing signal (1200 cycles). It has also been shown that the contact area dependent electrical response to applied pressure in TENGs can be utilised directly to measure contact pressure^[234-236].

highly stretchable and self-healing hydrogel-based sensor with exceptional stretchability, area expansion, and healing efficiency (**Figure 11a**). It had a gauge factor of 18.28, broad sensing range, lower detection limit, excellent thermal responsiveness (-0.0164 °C⁻¹), distance-dependent thermosensation (-0.022 °C mm⁻¹) and a high temperature resolution of 2.7 °C. Hao's team^[242] reported a strain and temperature dual-functional CPHs based WSSs by cooperating the thermosensitive TEMPO-oxidized cellulose/PANI networks in hydrophilic hydrogel polymer matrix through hydrogen bonds and electrostatic interactions (**Figure 11b**). They coupled the charge transfer caused by the spatial position and the tunneling current activated by thermal vibration to detect human motion and temperature simultaneously. By leveraging the superposed signals discrimination ability of CPs, the obtained CPHs exhibited excellent outstanding thermosensation (TCR = 2.01% °C⁻¹) and strain sensitivity (GF = 1.25).

Gas sensor

As gases are considered as biomarker of human diseases^[243-245], developing wearable gas sensors that can precisely detect specific gases at low concentrations has enormous value for human healthcare^[246-248]. PANI undergoes significant changes in its electric characteristics upon doping/dedoping with amines, making it an excellent material for the construction of gas sensors^[249]. For example, Wang et al.^[209] reported a room-temperature CeO₂@PANI hydrogel based NH₃ gas sensor, which showed a high response (6.5



to 50 ppm of NH₃) and short detection time (a response of 6.5 and a response time 57.6 s). The improved sensing performances can be attributed to the p–n heterojunction within the core–shell structure. Notably, the obtained CPHs sensor exhibited excellent long-term sensing stability, mainly due to the doping and crosslinking effects on the polymer chains. Furthermore, one of the greatest and least recognized risks to human is oxygen deprivation in confined places. Wearable oxygen sensors for detecting reduced oxygen level are urgently needed for protecting people from injuries and deaths^[250, 251]. Decataldo and co-workers^[211] developed a wearable oxygen sensor at room temperature based on PEDOT:PSS. The fast O₂ solubilization in the hydrogel allowed for gaseous oxygen transduction in an electrical signal thanks to the electrocatalytic activity of PEDOT:PSS. The obtained CPHs gas sensor showed a low power consumption of 30–40 μW.

Humidity sensor

Humidity is considered one of the most effective variables for respiration monitoring^[252], and high-performance humidity sensors has been given increasing concerns over the past years in the field of health assessment and disease prediction with their advantages of fast response, high sensitivity, wide monitoring range, low cost and simplicity.^[253, 254] PEDOT:PSS based CPHs are promising for sensing humidity that depends on water adsorption and desorption by the PSS. On one hand, the insulating and hydrophilic PSS shell is tending to absorb water and swell in high humidity conditions, thus increasing the space between adjacent PEDOT chains, resulting in a raised resistivity. On the other hand, low humidity results in water desorbing from the PSS, which reduces the PSS's volume and the space between neighboring PEDOTs, leading to a lower resistivity. Nowadays, PEDOT:PSS has been used to develop various types of humidity sensors^[207, 255, 256]. For example, Bian et al.^[208] developed an interpenetrating PEDOT: PSS based hydrogel humidity sensor with tunable mechanical properties and good conductivity. Thanks to the uniformed conductive component PEDOT:PSS and the hydrogen bonds between water molecules and the large amount of hydrophilic groups in the hydrogel, the obtained CPHs humidity sensor exhibited a wide relative humidity range (0–85%).

4.2.3 Other applications

Human-machine interfaces

The interactions of human-machine interfaces are crucial for mobile communications, intelligent robots, and intelligent medical care^[257]. Nowadays, increasing interests are considered to develop the flexible human-machine interactive devices based on CPHs with their advantage of soft, stretchable and biocompatible performance^[258]. Attributing to the ability of CPHs to convert human motion signals into electrical signals, electronic devices such as robots can clearly recognize the commands issued by the human movement and give corresponding feedback. For example, Jing's group^[259] developed a multifunctional CPHs. Besides fabricating the CPHs as strain and temperature sensor, the CPHs was assembled as a wearable human-machine interface device, and a smart glove was developed for hand gesture recognition by utilizing the exceptional CPHs, in order to transmit data to a smart phone real-timely via a bluetooth wireless system.

Implantable electrodes

For implantable electrodes, CPHs must have acceptable biocompatibility, which means the CPHs must have no major long-term effects in vivo or only cause a minor tissue response. For example, when PPy was implanted into the cerebral cortexes of rats, they were tolerated well and allowed the formation of complex neural networks^[260]. Nowadays, there are great prospects for using CPHs as implantable devices to monitor human's organ motions^[261]. For cardiac tissue engineering, Liu and co-workers^[262] reported the first functional heart patch that combined conductivity and spontaneous adhesion. The paintable CPHs was synthesized based on polymerizing pyrrole and dopamine in hyperbranched polymer chains. The obtained CPHs could be strongly bonded to the beating heart for 4 weeks, which efficiently boosts the transmission of electrophysiological signals. Consequently, there was a notable improvement in the revascularization of the infarct myocardium and the rebuilding of cardiac function. Notably, PEDOT:PSS based CPHs possessed both electronic and ionic conductivity with sufficient hydration. On one hand, the CPs chains are acting as the charge transfer pathway for the electronic carriers. On the other hand, ions are highly mobile in the water-rich hydrophilic polymer matrix^[263]. The unique ion/electronic mixed conductivity of PEDOT:PSS based CPHs endow them in a variety of bioelectronic interfaces^[264, 265].

Flexible energy storage system

CPHs can also be fabricated to work as flexible energy storage devices to power WSs in consecutive stretching, bending and twisting conditions^[266]. Principally, CPs inherently bond with the hydrophilic polymer matrix, constructing a continuous conducting and monolithic framework for facilitating the charge transportation. Moreover, an ideal interface between the electrode materials and electrolyte solution could be established by the porous architectures of the CPHs, promoting a quick and efficient electrochemical reaction^[267]. Consequently, CPHs are highly promising for energy storage devices such as supercapacitors and batteries to realize mechanical flexibility, ideal electrochemical performances and additional functions^[268, 269]. For example, Teng's group^[270] reported a high-performance fiber-shaped supercapacitor with high specific capacitance of 393.8 F cm⁻³ at 2 A cm⁻³ by in situ polymerization of PPy on PEDOT:PSS ECHs. The outstanding electrochemical performance of the fiber shaped ECHs supercapacitor can be attributed to the fast electrons transfer/ions diffusion and the efficient utilization of whole electrode, which lead to good electronic and ionic conductor properties, hierarchically porous structure of fiber shaped hydrogel and π–π interaction of CP network. The rate performance of 52.8% capacitance retention at ultrahigh current density of 50 A cm⁻³ probably owing to hierarchically conductive network and the highly porous interconnected nanostructures in the CPHs, which can accommodate the swelling and shrinking of the polymer network, facilitating the electronic and ionic transfer speed. Mi et al.^[271] synthesized an interconnected CPHs coated Si nanoparticles with reinforced conducting three-dimensional (3D) network structure and used as anode materials for lithium ion batteries. The synergy effect of CPHs enhances the electron transport capability, and results in excellent electrical, mechanical integrity and conductivity of the electrodes.

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REVIEW

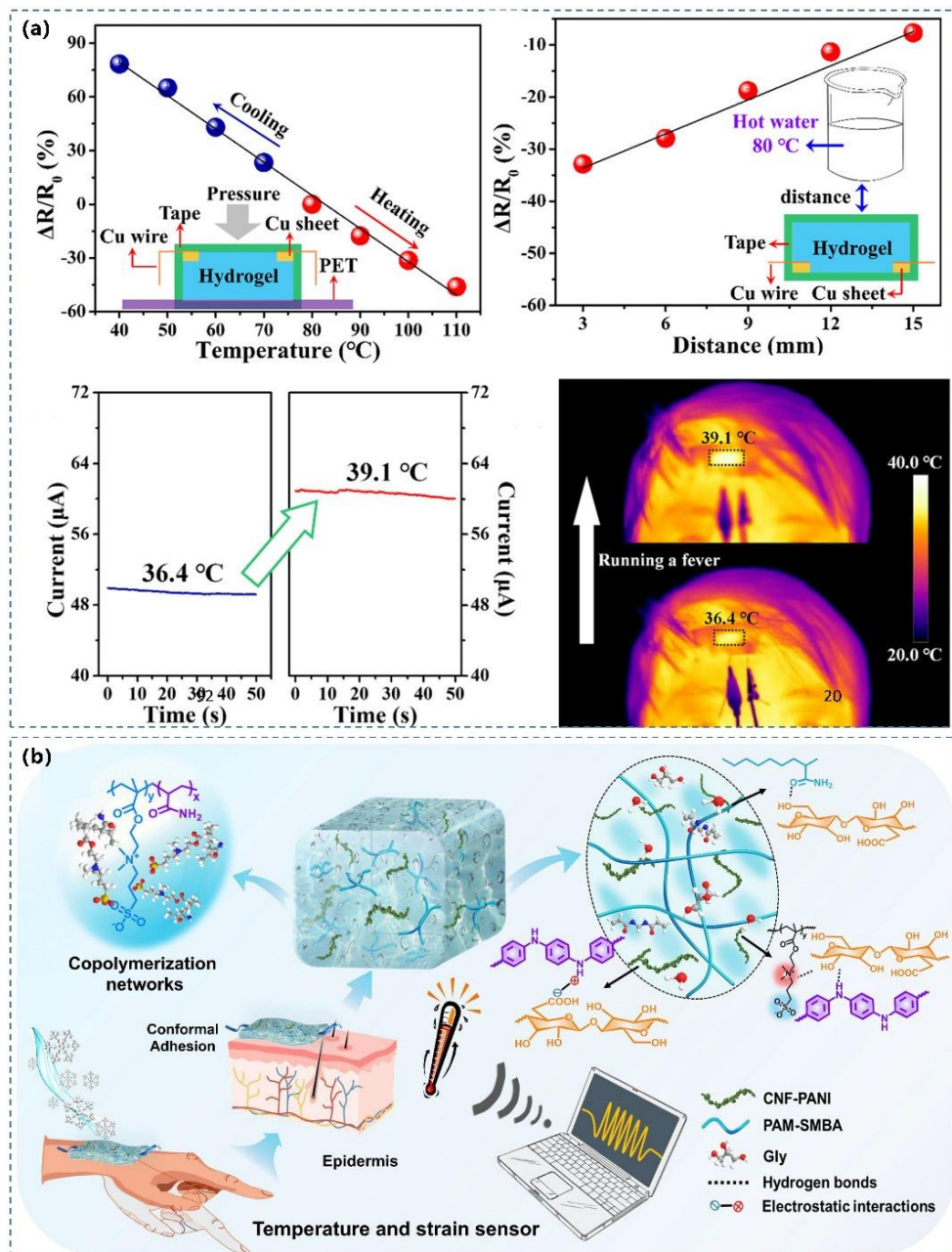


Figure 11. (a) Temperature sensor serves to monitor human forehead's temperature. Reprinted from ref. [22] with permission. Copyright 2020 American Chemical Society. (b) The design of PANI based hydrogel with strain and temperature responsiveness. Reprinted from ref. [242] with permission. Copyright 2022 Elsevier.

5. Conclusions and future prospects

In conclusion, the designability of CPHs and their structural closeness to natural soft tissues have sparked a lot of interest in converting

external inputs to electrical signals and monitoring human health in real time. The advantages of CPHs, including high conductivity, tunable mechanical characteristics, self-healing ability, self-adhesiveness, and adequate sensing sensitivity, make them a viable building block for various WSs. In this review, we first present the formation of molecular network structure, including single



component CPHs, CPs nanocomposites hydrogels and double-network CPHs. There are mainly two kinds of synthetic process to construct CPH that CPs either function as conductive fillers or create conductive frameworks. Secondly, mechanical characteristics, electrical conductivity, and other functionalities vital for WSS (such as self-healing, self-adhesiveness, and biocompatibility) are explored in depth. CPHs have made significant progress in pressure/strain sensors, temperature sensors, and temperature and pressure/strain sensors, thanks to their good electrochemical performance, tunable conductivity, and great mechanical qualities. New prospects and breakthroughs in the fields of medical monitoring and soft robotics will likely result from this recent progress.

Despite the reviewed progresses, there is still room for advancement in the field of conductive hydrogels. The fact that most reported CPHs exhibited a nonlinear dependence of sensitivity on low strain. It still remains challenges to obtain linearity, which is crucial for the viability and accuracy of CPHs based resistive typed-WSS. The stress transports from hydrogel matrix to conductive network during stretching, and the sensing signal is oriented from the change of conductive network. The linearity relies on reliable and continuous conductive network during deformations, which depends on stable and strong interactions between the conductive network and the flexible matrix. To improve the sensing linearity of the CPHs, a possible solution is to eliminate stiffness difference between CPs and hydrophilic polymer matrix by constructing chemical and physical chemical bonds between them with excellent affinity. As a result, the conductive pathways and the hydrogels could deform synchronously under strain, leading to a linearly sensing signal.

Another critical issue is the reliability and stability of CPHs based WSS in practical applications. The sensing stability of CPHs during long-term operation in biological environment is a key point for WSS. The evaporation of water would significantly disrupt the electrical conductivity and mechanical performance of CPHs, affecting their sensing functions. Introducing binary solvent such as glycerin or constructing hydrophobic organogel to fulfil the CPHs are efficient ways to improve the dehydration feature at an open-air, offering CPHs with long-term sensing stability under extreme environments such as high temperature or cold environment. Taking the appropriate designed CPHs a step further, they could monitor human motion under water without compromising their conductivity and mechanical properties. It is extremely desired to build specialized structures or incorporate organic agents to CPHs for reliable anti-freezing, long-lasting hydration, and long-term stability qualities. We believe that more trophies are to be collected by developing next generation CPHs based WSS, to improve the life quality of human being.

Conflicts of interest

There are no conflicts to declare.

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