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Conductive hydrogels for bioenergy harvesting and self-powered application

Chenyang Zhang [a,c,f,1], Md Osman Goni Nayeem [b,1], Zhiqi Wang [a,c,1], Xiong Pu [d], Canan Dagdeviren [b,*], Zhong Lin Wang [d,e,*], Xiaohong Zhang [a,c,*], Ruiyuan Liu [a,c,*]

[a] Soochow Institute of Energy and Material Innovations, Key Laboratory for Advanced Carbon Materials and Wearable Energy Technologies of Jiangsu Province, Institute of Functional Nano & Soft Materials (FUNSOM), College of Energy, Soochow University, Suzhou 215006, PR China
[b] Media Lab, Massachusetts Institute of Technology, Cambridge, MA 02139, United States
[c] Jiangsu Key Laboratory of Advanced Negative Carbon Technologies, Soochow University, Suzhou 215123, PR China
[d] CAS Center for Excellence in Nanoscience, Beijing Key Laboratory of Micro-nano Energy and Sensor, Beijing Institute of Nanoenergy and Nanosystems, Chinese Academy of Sciences, Beijing 100083, PR China
[e] School of Materials Science and Engineering, Georgia Institute of Technology, Atlanta, GA 30318, United States
[f] Research Institute for Intelligent Wearable Systems, The Hong Kong Polytechnic University Hung Hom, Kowloon 999077, Hong Kong, China

A B S T R A C T

Conductive hydrogels (CHs) are characterized by tissue-like mechanical properties, multiple stimuli-responsive abilities and favourable biocompatibility, enabling their wearable devices to construct a conformal interface with rugged tissue surfaces compatible with human body for real-time monitoring. However, external power supply requirements and complex fabrication processes limit their broad applications. Self-powered wearable devices can harvest energy from humans and the ambient environments, benefitting a long-term operating ecosystem. In this review, we discuss the materials engineering and designs for CHs-based devices for bioenergy harvesting and self-powered application. Specifically, we highlight the unique advantages of CHs in bioelectrical interfacing with the human body. CHs-based flexible bioenergy harvesters as well as self-powered applications are then summarized. Finally, we will provide the current challenges for materials, device designs and feasible approaches in the next-generation CHs-based multifunctional devices.

1. Introduction

Wearable electronics that can capture various signals from the human body are the most promising platforms for untethered healthcare-monitoring and human–machine intelligent ecosystems [1–4]. A wide range of flexible, nontoxic, and physiochemically tunable materials have been developed, aiming at overcoming the device-to-tissue mechanical mismatch from conventional rigid electronics [5,6]. Hydrogels, consisting of polymer networks resembling tissue structures and water molecules, can offer a wet environment for biological tissues and have long been used in biomedical applications [7,8]. For the application of wearable bioelectronics, the superior biocompatibility, stretchability, self-adhesion, and air-permeability make hydrogels perfect materials for seamlessly bridging human tissue and different rigid devices such as circuits, sensors and drug delivery systems [9]. By incorporating conductive materials such as conductive polymers [10,11], nanomaterials [12,13], and carbon-based materials [14,15] into the matrices, hydrogels could be conductive [16]. This is extremely important for bioelectronics that perform electronic communications

* Corresponding authors.
E-mail addresses: canand@media.mit.edu (C. Dagdeviren), zlwang@binn.cas.cn (Z.L. Wang), xiaohong_zhang@suda.edu.cn (X. Zhang), ryliu@suda.edu.cn (R. Liu).
[1] These authors contributed equally to this paper.

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between biology and electronics. On the other hand, conventional electronics rely on external power supplies and complex circuit design for the ecosystem running, limiting the activity of wearers [17,18]. CHs have also been extensively employed as substrate, electrodes, or active materials for energy conversion and sensing applications [19,20]. Considering these factors, a CH-based self-powered stretchable and flexible sensing system that harvests bioenergy and actively generate signals can enable a wearable system for wirelessly long-term monitoring and real-time human–machine interaction [21,22].

Bioenergy exists in various forms, such as mechanical energy from human motion [23], water resources and biofluids from sweat and environments [24], and thermal energy from temperature gradient between human body and the surroundings [25], which can be converted to electricity energy by triboelectric nanogenerators (TENGs) [26,27], piezoelectric nanogenerators (PENGs) [28,29], hydrovoltaics [30,31], thermoelectric generators (TEGs) [32,33], and biofuel cells (BFs) [34,35]. As electrodes and substrates for stretchable and flexible TENGs and PENGs, CHs possess unique advantages in the superb bio-adhesion with the curvature of skin or organs [36], which can guarantee effective energy harvesting and accurate signal detecting from even tiny motions [37–39]. Multi-functional hydrogels can easily absorb liquid water or moisture, provide continuous water resource for hydrovoltaic devices with their high-porosity, and produce electrical power directly from the coupling between active materials and assorted forms of water [40]. Hygroscopic hydrogel coolers have also been developed for effective heat removal [41]. The performance of CHs-based devices for bioenergy harvesting and self-powered sensing have been gradually improved [42], while there still exist challenges and room in materials engineering and device design for the real-world applications [43]. One of the challenges faced by current CHs-based bioenergy harvesters is that they are not able to provide enough power for daily used portable electronics such as smartwatches and cellphones [24]. Numerous studies focus on improving the mechanical performance of CHs to meet the physical demands during energy harvesting and the water retention abilities to maintain their long-term functionalities [44–46]. Nevertheless, the harsh environments in which these devices are possibly used can result in accelerated degradation of the hydrogels, which will decrease both the stability and electrical output of the devices [47,48]. For materials engineering, multi-functionalization can meet the requirements of harsh environments or particular applications. In the part of device design, integration strategy and power management are essential for more efficient energy harvesting, storage, and utilization.

In this review, we discuss the state-of-art CHs-based stretchable and flexible devices for bioenergy harvesting and self-powered applications, including materials engineering, device designs and applications (Fig. 1). Firstly, we introduce principles and synthesis of different conducive types of CHs. Important properties and materials engineering are then discussed. Device designs and performance of bioenergy harvesters are presented, following with the self-powered applications. Finally, current challenges and opportunities for materials engineering and devices design for real-world applications are proposed, providing a roadmap for future developments in this field.

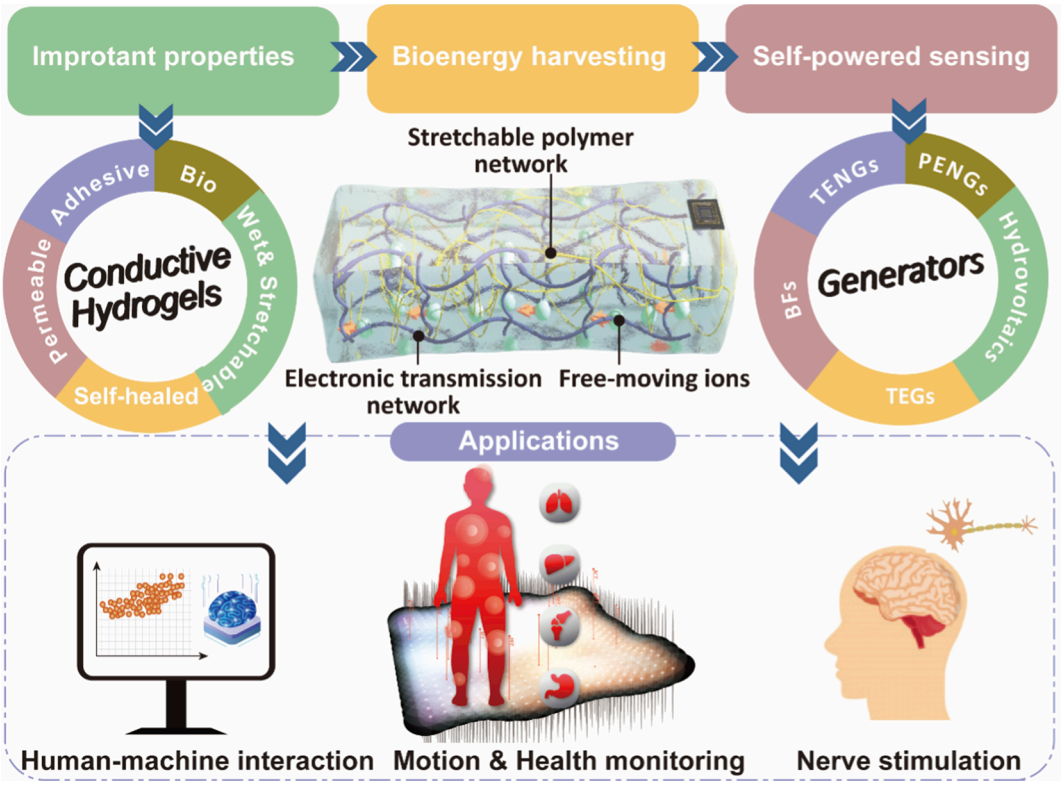

Fig. 1. The introduction of recent CHs with mechanism, applications on bioenergy generating, and self-powered applications. Triboelectric nanogenerators (TENGs), piezoelectric nanogenerators (PENGs), thermoelectric generators (TEGs), biofuel cells (BFs).

2. Conducting principles and synthesis methods

CHs prepared from various types of conducting materials by different synthesis methods can form different conductive pathways, which have important effects on energy transfer, signal interaction sensing, reaction sensitivity, and reaction stability. Therefore, it is necessary to understand the conductive mechanism and synthesis method of CHs in order to design desirable materials for various applications. Based on the conducting mechanisms, CHs can be divided into three types: electron-CHs, ion-CHs, and electron–ion composite CHs. Among them, electron-CHs are mainly conductive by the migration of electrons, the conductivity of ion-CHs is caused by free ions or ionized groups in hydrogel network, and the conductivity of ion–electron type CHs is generated by the simultaneous migration of electrons and ions. The properties and applications comparison of different types of CHs have been conducted and summarized in Table 1, the conductivity, content, miscibility, toxicity and biocompatibility of common conductive materials have been summarized in Table 2.

2.1. Conductive hydrogels

The development of conductive hydrogels has evolved over time. The research of hydrogels began in the 1960s, focusing on their use as biocompatible materials [49]. In the past decade, researchers began incorporating conductive components into hydrogels to impart electrical conductivity, including conductive polymers and nanoparticles [50,51]. As technology advanced, CHs with tailored properties emerged for biomedical applications. By mimicking the hierarchically organized structure of spider silk, researchers have synthesized high performance stretchable conductive hydrogel [52]. Moreover, the conductive properties of the heart tissues, particularly the cardiac conduction system, have inspired the development of conductive hydrogels for engineering cardiac tissues, creating bioelectrodes, and studying heart-related diseases [53,54]. For bio-applications, CHs-based bioelectronics enable the delivery of electrical signals to tissues or cells, which is crucial for applications like neural stimulation [55], tissue engineering [56], and regenerative medicine [57]. The nervous system in humans and animals relies on electrical impulses for communication between

Table 1
. Properties and applications comparison of different types of CHs.

Components of polymer networks	Conductive material	Conductivity (S/cm)	Polymer type	Conductivity type	Application	Ref.
[P(PEG-*co*-AA)]/PANI	PANI	$7.43*10^{-2}$	conductive polymers-based	electronic type	flexible sensors	[58]
PPy−PAM/CS	PPy	$3*10^{-3}$	conductive polymers-based	electronic type	wound dressings/sensors	[68]
Gel/HA/PEDOT:LS	PEDOT	$6.9*10^{-3}$	conductive polymers-based	electronic type	biomimetic scaffolds	[90]
PEDOT:PSS	PEDOT:PSS	40	conductive polymers-based	electronic type	bioelectronic device fabrication	[91]
TOCNF-Graphene/PAA	Graphene	$2.5*10^{-2}$	carbon-based	electronic type	wearable monitoring of human motion	[89]
PTCM-Gly	MXene	$1.1*10^{-3}$	carbon-based	electronic type	motion monitoring	[92]
MWNT-gelatin	MWNT	$1.6*10^{-3}$	carbon-based	electronic type	robotic skin	[93]
PSGO-PEDOT-PAM	PSGO/PEDOT	8.297	conductive polymers/ carbon-based	electronic type	bioelectronics	[73]
AgNP/MeHA-Ga	AgNP	$5*10^{-2}$	metal-based	electronic type	3D printing of lattices/bridge muscle defects.	[94]
Ag-PAAm–alginate	Ag	350	metal-based	electronic type	wearable electronics	[74]
$Li^+/Ca^{2+}/K^+$-TOCNF	$Li^+/Ca^{2+}/K^+$	$4.36*10^{-4}$	free ion	ionic type	artificial intelligence products	[95]
PVA-CNF	Na^+/Cl^-	$3.2*10^{-2}$	free ion	ionic type	multi-functional sensors	[96]
PVA/PEI/Li^+	Li^+	11.76	free ion	ionic type	artificial nerve fiber	[76]
PUU/[EMIm][DCA] IL	([EMIm][DCA])	$2.25*10^{-2}$	ionic liquid	ionic type	3D printable	[97]
PVC/DBA/[$EMIM^+$] [$TFSI^-$] IL	[$EMIM^+$] [$TFSI^-$]	$1.37*10^{-5}$	ionic liquid	ionic type	monitors human motion	[98]
CNF/PAM wood hydrogel	CNF	$5*10^{-4}$	ionic polymer	ionic type	biomaterials/ nanofluidic	[83]
HBA−AMPS−graphene	AMPS/graphene	$4.87*10^{-2}$	ionic polymer/ carbon-based	electronic/ionic type	wearable devices	[99]
PPy-PAA-Fe^{3+}	PPy/Fe^{3+}	$1.4*10^{-1}$	free ion/ conductive polymers-based	electronic/ionic type	smart energy storage devices	[87]

polyethylene glycol (PEG); acrylic acid (AA); polyaniline (PANI); polypyrrole (PPy); acrylamide (AM); chitosan (CS); gelatin methacrylate (Gel); hyaluronic acid methacrylate (HA); poly(3, 4-ethylenedioxythiophene): sulfonated lignin (PEDOT:LS); poly(3,4-ethylenedioxythiophene): sulfonated lignin (PEDOT:PSS); TEMPO-oxidized cellulose (TOCNF); polyacrylamide-tannic acid-decorated cellulose nanofibrils- Mxene (PTCM); glycerol (Gly); multi-walled carbon nanotubes (MWNT); polydopamine-reduced and sulfonated graphene oxide (PSGO); silver nanoparticles (AgNP); The hyaluronic acid was modified with both methacrylates and gallols (MeHA-Ga); polyvinyl alcohol (PVA); polyethyleneimine (PEI); poly(urethane-urea) (PUU); methylimidazolium dicyanamide ([EMIm] [DCA]); PVC: polyvinylchloride; dibutyl adipate (DBA); bis(trifluoromethylsulfonyl) imide ([$EMIM^+$] [$TFSI^-$]); 4-hydroxybutyl acrylate (HBA); 2-Acrylamido-2methylpropane sulfonic acid (AMPS).

Table.2
Conductivity, content, miscibility, toxicity and biocompatibility of common conductive materials.

Conductive materials	Conductivity (s/m)	Content	Miscibility	Toxicity	Biocompatibility	Ref
CNTs	0.05–30	0.01%-1%	poor	weak	good	[100–103]
GO	$1.0*10^{-4}$-110	0.01%-2%	poor	very weak	excellent	[69,73,89]
Mxene	$5*10^{-4}$-10	0.001%-0.2%	poor	very weak	excellent	[72,92,104]
PANI	0.1–10	0.1%-0.5%	good	weak	good	[58,59,105]
PPy	0.1–15	0.1%-20%	poor	weak	good	[60,61,68]
PEDOT:PSS	$300–4*10^{4}$	30%-100%	good	weak	excellent	[63,64,91]
metal-based materials	$1.0–3.5*10^{4}$	5%-50%	poor	weak	good	[74,75,106]
free ion	0.1–1000	1%-30%	good	very weak	excellent	[76,77]
ionic liquid	0.01–1.0	5%-40%	good	very weak	excellent	[78,79,97]
ionic polymer	$1.0*10^{-2}$-15	0.1%-10%	good	very weak	excellent	[80,81,95]

neurons. Furthermore, CHs can mimic the electrical conductivity of native tissues and the structure of extracellular matrix, making them suitable for tissue engineering. For instance, cardiac tissue engineering requires materials that can support the propagation of electrical signals, similar to the specialized conductive system in the heart. CHs can also be used as bioelectrodes for sensing and monitoring biological signals. They can be integrated with biological systems to measure neural activity, muscle contractions, and heart rhythms.

2.2. Electronic type

2.2.1. Conductive polymers

Conductive polymers are a kind of polymers with easy synthesis, simple structure, high electrical conductivity and adjustable physicochemical properties, such as polyaniline (PANI) [58,59], polypyrrole (PPy) [60,61], polythiophene (PTh) [62], poly[3,4-(ethylenedioxy)thiophene](PEDOT) [63–65], and polyacetylene [66]. Conductive polymers have continuous sp^2-hybridized carbon-centered backbones containing alternating single and double bonds arranged in a π-π conjugated structure. The loss of an electron in the conjugated π orbital under the effect of electron delocalization forms the delocalized electron. The electron delocalization can move freely in the polymer backbone, resulting in electron motion that gives the polymer its conductive properties. Polymeric CHs with excellent electrical conductivity are prepared by compounding conductive polymers with natural polymers. Currently, there are two main methods for the preparation of polymeric CHs, one is to compound conductive polymer with a natural polymer/monomer with excellent hydrophilicity and biocompatibility by self-assembly or introduction of a cross-linking medium. In this case, the conductivity of the hydrogel can be optimized by regulating the content of the conducting polymer. For example, a novel γ-polyglutamic acid (γ-PGA)/PEDOT:PSS CHs with excellent conductivity (~12.5 S/m), adhesion and self-healing properties has been prepared [67]. The electrical conductivity, self-healing properties, and adhesion of the hydrogels can be adjusted by varying the amount of PEDOT:PSS in γ-PGA/PEDOT:PSS hydrogels. The sensors prepared using hydrogels are designed to be able to monitor different human behaviours through the change of electrical signals during deformation. Schematic diagram of the structure of (γ-PGA)/PEDOT:PSS hydrogels is shown in Fig. 2A.

Another approach is to prepare CHs by chemical oxidation, in-situ polymerization or growing conductive polymers by electrochemical polymerization. Controlling the content of conducting polymers through different conditions enables the adjustment of the electrical conductivity and overall performance of the hydrogel. A polymeric CH with excellent electrical and mechanical properties as well as good biocompatibility has been composited by in situ polymerization of PPy nanorods under the condition of chitosan backbone as a biotemplate [68]. The mechanical properties and electrical conductivity of the prepared hydrogels can be tuned by adjusting the content of chitosan in the pre-hydrogels and the concentration of PPy nanorods.

2.2.2. Carbon-based materials and MXene

There are various types of conductive carbon materials, such as graphene, graphene oxide (GO), carbon nanotubes (CNT), and Mxene, which have excellent electrical conductivity, very high specific surface area, excellent mechanical strength [69–72]. These carbon materials have high π-conjugated molecular chains, which facilitate the formation of delocalized electrons that can move more easily between atoms. The distribution of conductive carbon materials as conductive fillers in the polymer matrix enables the preparation of CHs with excellent properties. When the filling content of conductive carbon material increases to a critical value, three-dimensional conductive channels will be formed in the hydrogel, so that the conductivity is greatly enhanced. And when the conductive carbon material content is lower than the critical value, the conductive particle spacing increases and cannot form a conductive channel, which still has a low conductivity phenomenon at this time, mainly because the thermal vibration electrons inside the hydrogel are able to migrate between the conductive particles. In recent years, the preparation methods of CHs based on carbon materials are mainly divided into two types.

The first one is to obtain composite CHs by dispersing nanostructured carbon materials in hydrogel precursor solutions and polymerizing them. The first method is prepared by polymerization of nanostructured carbon materials dispersed in a hydrogel precursor solution. For illustration, CHs with excellent electrical conductivity, adhesion, and biocompatibility have been prepared by self-assembly of PEDOT on polydopamine-reduced and sulfonated graphene oxide template [73]. The modification significantly enhanced the electrical conductivity and hydrophilicity of graphene, which led to the enhancement of the electrical conductivity as

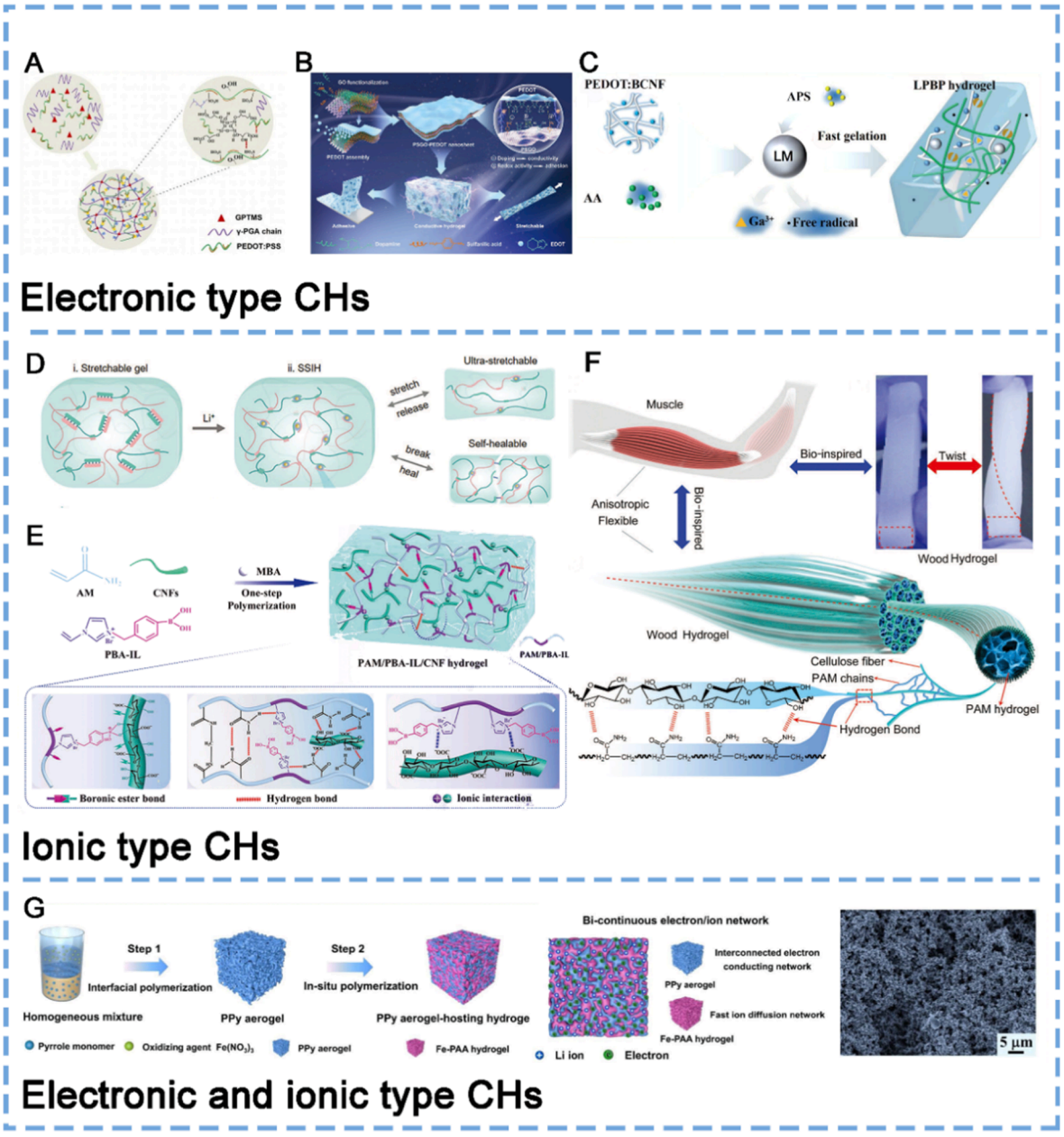

Fig. 2. (**A**) Schematic diagram of the structure of (γ-PGA)/PEDOT:PSS hydrogels. Reproduced from ref. [67] with permission from Elsevier Ltd, copyright 2022. (**B**) Schematic diagram of hydrophilic, conductive, and redox-active sandwich-like PSGO-PEDOT nanosheet and its incorporation in CHs. Reproduced from ref. [73] with permission from Wiley-VCH, copyright 2020. (**C**) Schematic of the gelation mechanism of liquid metal nanoparticle CHs. Reproduced from ref. [75] with permission from Royal Society of Chemistry, copyright 2021. (**D**) The design process of Li^+ self-healing hydrogel and the principle of self-healing ability and ultra-stretchability. Reproduced from ref. [76] with permission from Wiley-VCH, copyright 2022. (**E**) Fabrication mechanism and hierarchical dynamic networks in the construction of the PAM/PBA-IL/CNF CHs. Reproduced from ref. [78] with permission from Wiley-VCH, copyright 2022. (**F**) Schematic illustration and network microscopic structure of wood hydrogel. Reproduced from ref. [83] with permission from Wiley-VCH, copyright 2018. (**G**) Fabrication, schematic and SEM image of the host–guest hydrogels. Reproduced from ref. [87] with permission from Elsevier Ltd, copyright 2022.

well as the mechanical properties of the prepared hydrogels. The stretchable conductive hydrogel can measure the change of current during deformation and realize the monitoring of human movement status. The structure schematic and properties of the prepared hydrogel are shown in Fig. 2B.

Another method is to pre-synthesize the carbon material framework and then infiltrate the hydrogel pre-polymerization solution into the carbon material support for polymerization. For instance, conductive fillers such as Mxene, GO, and CNT on tetrapolar-shaped

zinc oxide (t-ZnO) templates with a mesoporous 3D network structure have been covered by drop-coating. The thickness of the generated nanomaterial layer was regulated by adjusting the dispersion concentration and repeating the infiltration process several times. Subsequently, the hydrogel precursor solution was permeated throughout the template to insert the hydrogel. After polymerization of the hydrogel, a hollow microchannel hydrogel containing MXene, GO, or CNT was prepared by wet chemical method to remove the t-ZnO template [69].

2.2.3. Metal-based materials

The classical theory of electrical conductivity suggests that there is a large number of free electrons that can move freely inside a metal conductor, and that the electric field force causes their directional movement to form an electric current, giving the metal excellent electrical conductivity. Liquid metal nanoparticles, metal nanowires (such as silver nanowires), metal nanoparticles (such as gold nanoparticles), and metal nanorods with low-dimensional nanostructures have high electrical conductivity, as well as excellent optical properties and are easy to process and modify, making them suitable candidate materials for electronic CHs. Therefore, the preparation of composite hydrogels with excellent properties by compounding nanostructured metallic materials with polymers is an intuitive and effective way to enhance the electrical conductivity. The addition of metal nanoparticles, liquid metal suspensions into the hydrogel precursor solution for direct gelation or physical embedding of metal nanoparticles in the pre-hydrogel matrix is a common method. In this approach, most of the conductive metals are used only as conductive components, and stable polymer chains are needed to provide structural support. A CH composite with high electrical conductivity, low Young's modulus, and high tensile properties by embedding silver sheets in a polyacrylamide-sodium alginate hydrogel, polymerizing and then removing the appropriate amount of water to induce permeation and form conductive channels [74]. A key part of the preparation process is the dehydration of the hydrogel, which promotes close contact between the silver sheets and keeps the permeation network formed intact and stable. In addition, the formation of reactive metal nanoparticles in pre-hydrogels or the cross-linking of metal nanoparticles with polymers can also lead to excellent electrical conductivity. Here, metal nanoparticles not only act as conductive components, but also participate in the formation of hydrogel networks to further improve conductivity and mechanical properties. A novel all-polymer CH based on liquid metal nanoparticles activated and cross-linked polyacrylic acid backbone has been composited [75]. The schematic diagram of the gelation mechanism for the preparation of CHs is shown in Fig. 2C. Liquid metal particles as cross-linking agents promote gelation in a much shorter time than conventional thermal or UV initiation processes, and the resulting CHs have excellent electrical conductivity, super-stretchability, and rapid self-healing properties.

2.3. Ionic type

2.3.1. Free ion (Electrolyte)

The hydrogel contains a large amount of water, which provides a good environment for dissolving acids or salts and forming abundant free ionic groups. The directional migration of these free ions generates ionic currents that provide the hydrogel with high electrical conductivity. The free ions in hydrogels are mainly divided into two categories, one is simply in the form of free ions in the hydrogel network, without any interaction with other components in the hydrogel [20]. The other involves with the ionic complexation or electrostatic interactions with polymer monomers and networks, which introduces ionic conductivity into the hydrogel while participating in the construction of the hydrogel network structure, enhancing the mechanical properties, porous network structure, and flexibility of the hydrogel. Alternatively, the CHs can be prepared by immersing the pre-prepared hydrogel in a certain concentration of acid or salt solution to displace free ions into the hydrogel network.

As shown in Fig. 2D, Li^+ has been introduced into the mixed hydrogel precursor solution of amino-rich polyethyleneimine and polyvinyl alcohol to form a network structure of hydrogel based on hydrogen bonding and electrostatic interactions, and construct a functional hydrogel with excellent electrical conductivity, self-healing properties, and hyperstretchability [76]. Changing the amount of lithium chloride in the hydrogel enables adjustment of the conductivity of the hydrogel. At the same time, the prepared hydrogel has reliable, high throughput information interaction and long-term stable operating characteristics, which is expected to work as a communication unit for information. A Zn^{2+}/sodium alginate based super-stretchable ion-conducting double-network (DN) polymer hydrogel has been designed [77]. Based on the framework structure of sodium alginate, functional monomers acrylic acid and acrylamide were introduced to build a DN structure, which enhanced the mechanical properties and provided more channels for ion migration. The amphiphilic structure of the polymer chains in the hydrogel can combine with Zn^{2+} to control ion migration and promote electrical conductivity.

2.3.2. Ionic liquid

Ionic liquids are molten organic salts, usually composed of anionic and cationic pairs of organic ions and inorganic counterions. The anions and cations of these compounds differ greatly in volume, and the asymmetry of certain substituents in the structure prevents the ions from accumulating regularly, with a loose structure and small forces between the anions and cations, making them easy to separate. Therefore, ionic liquids with good ionic conductivity, electrochemical stability and wide electrochemical window are a kind of promising material for the preparation of CHs. A novel method for the preparation of CHs has been proposed by designing a phenylboronic acid ionic liquid (PBA-IL) with multiple functions [78]. CHs with semi-interpenetrating networks were fabricated by introducing nanocellulose fibers (CNF) into the PBA-IL/acrylamide crosslinking network. Fabrication mechanism and hierarchical dynamic networks in the construction of the CHs are shown in Fig. 2E. Due to the synergistic effect of dynamic covalent and non-covalent bonds, hydrogels exhibit excellent self-healing properties, good mechanical strength, high adhesion, and excellent electrical conductivity, which have potential applications in the field of wearable bioelectronics. A series of multifunctional ion-CHs based

on choline-amino acid polyionic liquids (Cho-AA PILs) based on a DN approach has been designed [79]. Based on the reversibility of metal–ligand bonds, hydrogen bonds and electrostatic interactions in hydrogels, the synthesized hydrogels have efficient self-healing ability, excellent electrical conductivity, and antibacterial properties, and are capable of monitoring large human movements and minute physiological activities, with promising applications in the fields of artificial intelligence and biomedicine.

2.3.3. Ionic polymer

In addition to free ions and ionic liquids, polymers that partially contain ionized groups can also be a source of ions in hydrogels. These polymers, such as sodium carboxymethyl cellulose, 2-acrylamido-2-methylpropanesulfonic acid, and polyimide, can produce freely movable ions during hydrolysis, which will undergo a certain degree of directional diffusive motion in the polymer network under applied electric fields and have a certain degree of electrical conductivity [80–82]. These polymers are also involved in the construction of the hydrogel network, enhancing the overall performance of the hydrogel. Inspired by the muscle arrangement structure, a wood hydrogel with excellent ionic conductivity and anisotropy has been developed by exploiting various anisotropic structural and mechanical properties of natural wood and the flexibility of hydrogel [83]. CNF with negative charge-oriented arrangement in the wood forms nano-conductive channels inside the hydrogel, giving the hydrogel a high ionic conductivity of 5 $\times$ 10^{-4} S/cm. And the stronger bonding and cross-linking between CNF and acrylamide polymer brings better longitudinal properties. Schematic illustration and network microscopic structure of wood hydrogels are shown in Fig. 2F.

2.4. Electronic and ionic type

It has found some conductive polymers can form a strong mutual synergy with some ions, due to the presence of a large number of free ions and electrons inside the electron–ion composite CHs network, showing excellent electronic and ionic conductivity, relative to electron-CHs and ion-CHs, based on the interaction between ions and polymers, giving the hydrogel unique mechanical properties, flexibility and multi-functionality, has attracted increased attention in recent years [84,85].

A DN hydrogel with a combination of physically cross-linked GO and chemically cross-linked PAA has been constructed by compounding GO, AA and $CaCl_2$, with Ca^{2+} introduced into the hydrogel network as a dynamically reversible cross-linking agent, bridging neighboring GO as well as PAA and GO [86]. The prepared hydrogels were reduced to GO by hydroiodic acid vapor, and the

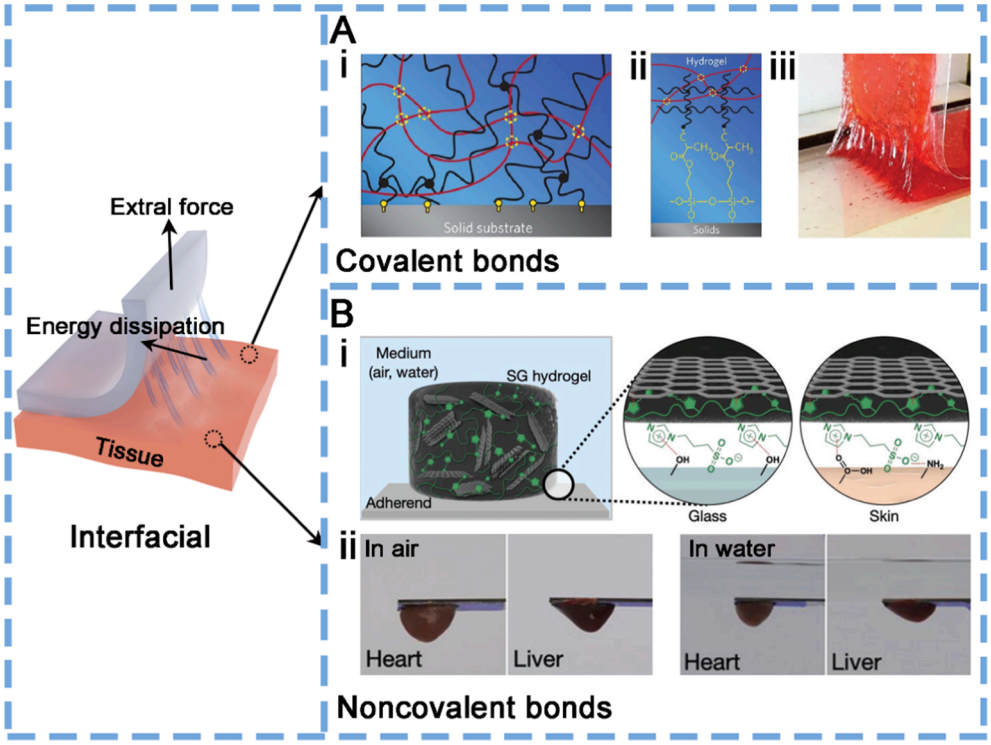

Fig. 3. Interfacial adhesion of CHs. (**A**) Covalent bonds. i) The tough bonding requires high fracture toughness of the constituent hydrogels. ii) The chemical anchoring of long-chain polymer networks is achieved by crosslinking the networks to functional silanes grafted on the surfaces of various solids. iii) Photo of the peeling of a tough hydrogel. Reproduced from ref. [121] with permission from Springer Nature Limited, copyright 2016. (**B**) Noncovalent bonds. i) The possible adhesion mechanisms of charge interactions between SG hydrogels and the adherends (glass and skin). ii) Photographs of adhesive property of hydrogel in air and in water with tissues. Reproduced from ref. [128] with permission from Wiley-VCH, copyright 2023.

resistance decreased sharply from 1473.23 kΩ for original samples to 257.31 kΩ, which slightly improved its conductivity. At the same time, the prepared hydrogels also exhibit extraordinary mechanical strength and self-healing efficiency. As shown in Fig. 2G, conducting polymer host–guest hydrogel electrodes with bi-continuous electron/ion transport has been prepared by promoting in situ polymerization of Fe^{3+}-liganded PAA hydrogel pre-solution in a constructed PPy aerogel conductive network [87]. The bi-continuous conductive network structure consists of electronically conducting PPy aerogel and ion-conducting Fe-PAA hydrogel, which utilizes the non-covalent cross-linking of hydrogen bonds and the reversibility of metal coordination to confer high electrical conductivity, superelasticity, excellent capacitance and inherent self-healing properties to the hydrogel electrodes, providing a promising strategy for the preparation of flexible all-solid-state supercapacitors. In addition, some ionic conductive polymers and electronic conductive polymers can directly undergo physicochemical cross-linking reactions to co-construct the network structure of the hydrogel, forming an electronic-ionic composite CH with excellent tensile properties and electrical conductivity performance [88,89].

3. Important properties and design strategies

Along with controllable physicochemical properties, CHs have been adopted in extensive applications in bioelectronics [107,108]. Unlike dry and abiotic materials, the unique tissue-like microstructure and water content endow a processing interface with various devices and human bodies [36,95]. However, CHs have their own limitations, including poor environmental stability, unexpected damages, and poor air permeability. Aiming for long-term, non-invasive bio-signals capture and continuous bioenergy harvesting, elaborately designed multifunctional CHs with adapted stretchability [109], tissue-like self-healing [110], and good air permeability [106] have attracted considerable interest. In this section, we review the current fundamental properties of CHs as wearable bioelectronics and the main design strategies to synthesize them.

3.1. Interfacial adhesion

As an alternative to conventional rigid electronics, existing devices, such as polymerics, textiles, and elastomers as substrates, have been employed to communicate and interact with human bodies [111–113]. However, tissue bumpiness and wet nature contradict these dry and abiotic materials. Air pockets and microscale gaps between human bodies and these flexible devices are inevitable during human daily activities, preventing the formation of ideal physical contacts, restricting the precise monitoring of bio-signals as well as the efficiency of bioenergy harvesting [114]. Meanwhile, conventional electronics mostly rely on physical attachments that face the challenges of unstable adhesion or tissue allergic reactions [115]. In contrast, CHs with great interfacial adhesion can offer direct and tight body contact and have been used in various tissue-interfaced bio-applications, such as health tracking [116], wound care [117], human–machine interaction [118], and bio-adhesive ultrasound for continuous imaging of internal organs [119] without extra binders (Fig. 3).

Composition is a crucial factor when considering the interfacial adhesion of CHs, which consist of water, polymers, and conductive materials. These different components play a major role in reducing or enhancing their interfacial adhesion [8,36]. The precise nature of its adhesion depends on the topology of connection, mechanics of dissipation, and the interaction between the components [120]. The most straightforward way for CHs to achieve adhesion with different interfaces is using covalent bonds, which can result in the chemistry linking between the polymer chains and the targeted surfaces (Fig. 3Ai) [121,122]. Covalent bonds, with two or more atoms sharing their outer electrons to form a relatively stable chemical structure, is much stronger than other adhesive bonds. Whereas surface modification and suitable polymer and crosslinkers selection is needed [115,123]. A tough CHs covalently to non-pours solids interface through the silanation between each surface has been achieved, leading to a high interfacial toughness of over 1,000 J/m^2 (Fig. 3Aii-iii). Ionic interaction is another covalent bond for adhesive CHs, such as ionic CHs and/or metal oxide surfaces. These covalent can make a reversible interaction for CHs adhesion. However, the specific surface requirements for ionic covalent bonds make them hard to cooperate with other types of bonds for CHs interfacial adhesion strategies. Covalent bonding is typically strong but irreversible, which makes it unsuitable for repeated adhesiveness [78,124].

Instead, various reversible noncovalent bonding mechanisms are commonly used to achieve repeatable hydrogel adhesion. CHs adhere through non-covalent interaction involving the construction of networks in the interfaces via one or a combination of physical interactions, including van der Waals interaction, electrostatic attraction, π-π/cation-π interaction, ion/dipole–dipole, and hydrogen bonding (Fig. 3Bi). Van der Waals interaction and electrostatic attraction are the most widely uased strategies. However, the nature of CHs (water-filled) will significantly weaken these two interactions. For repeatable adhesion and peeling with different surfaces, π-π/cation-π interaction, ion/dipole–dipole and hydrogen bonding have been widely used. In general, non-covalent bonds are weaker than covalent bonds. For immediate and repeatable adhesion and peeling CHs adhesion, various non-covalent interactions are cooperating together to achieve energy dissipation during peering process for highly adhesive CHs [73,78,125]. Meanwhile, water is inherently capable of hydrogen bonding and various polymer chains inside CHs (PDA, Chitosan, poly (SBVI) *etc.*) can maintain abundant noncovalent bonding groups between the CHs and different substrates, causing high interfacial adhesion (Fig. 3Bii) [126–128]. Furthermore only a few adhesions between cationic gels and anionic gels are based on electrostatic attraction [129].

Adhesive CHs have held great processes in multiple bioelectronic applications that can attach to human tissue directly. For CHs-based TENGs, the interfacial bonding between elastomers and CHs is weak, which is easy to cause crack in triboelectric layer. It has been shown that adhesive hydrogel can enhance adhesion between the elastomer and CHs for more efficient energy harvesting [43]. For epidermal sensors, CHs with good adhesion can acquire more precise data for health monitoring. Despite these advantages, it is still challenging for the strong adhesion in a damp environment. When the CHs adhere with a hydrophilic surface, water molecules will form a water film, preventing the contact between the substrate and CHs [130]. Serval approaches, such as hydrophobic polymers and

catechol group chemistry, have been inspired by mussels' adhesion mechanism [131–133]. For example, a type of underwater adhesion with human skin and various substrates has been achieved by utilizing the hydrophobic anion within CHs to disrupt the hydration-layer barriers of the substrate [134].

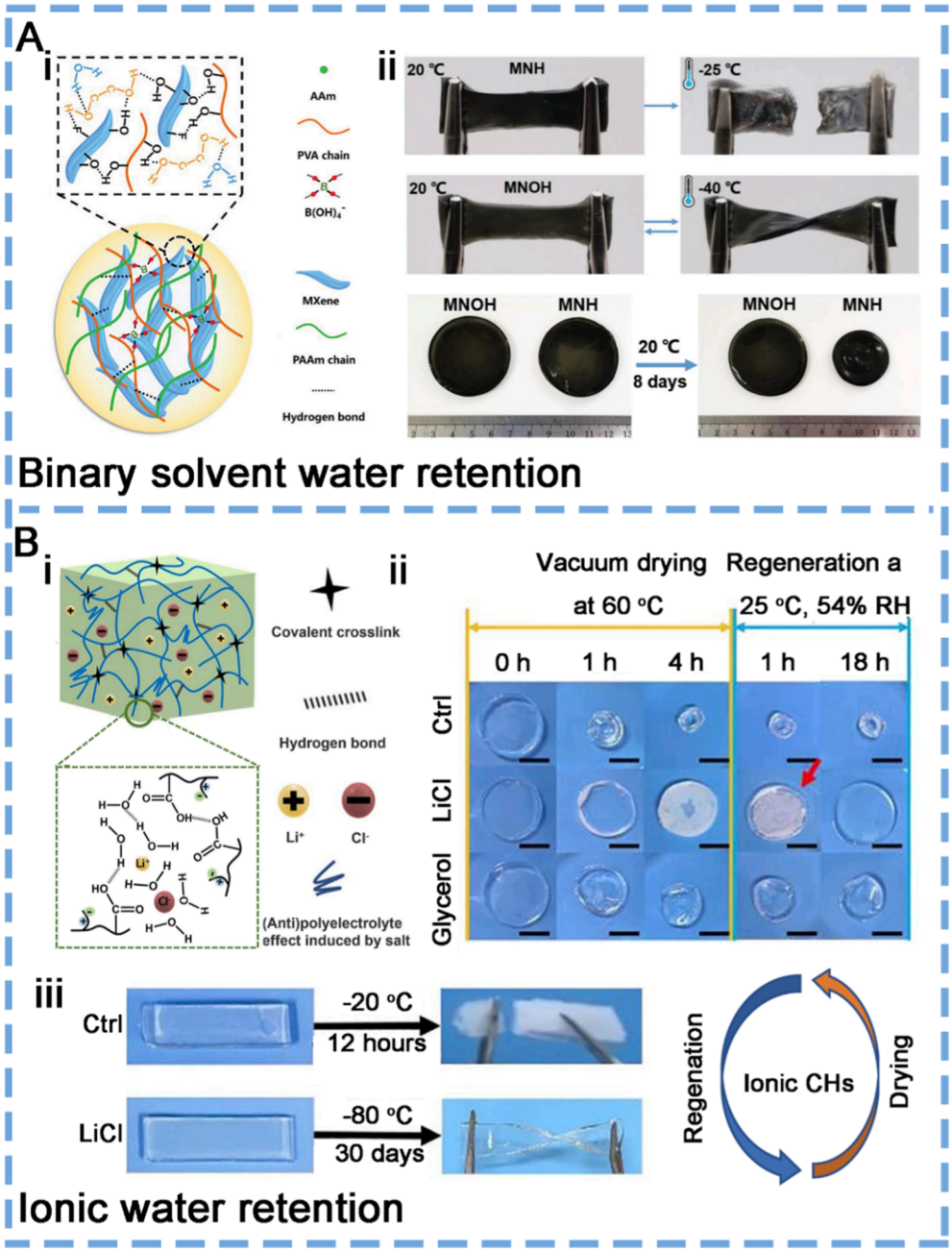

Fig. 4. Water retention ability. (**A**) Binary solvent anti-freezing CHs. i) Schematic illustration of the fabrication of a CH with good water retention ability. ii) Photographs of anti-freezing and long-lasting moisture retention performance. Reproduced from ref. [141] with permission from Springer Nature Limited, copyright 2021. (**B**) Ion dropping water retention CHs. i) Images of control, glycerol-based, LiCl-based hydrogels stored at 25 ℃ and 54% RH for 0 and 7 days. ii) Compressive modulus of original LiCl-based hydrogels and the ones stored at 25 ℃ and 54% RH for 7 days. iii) Twist of control or 30% LiCl-based hydrogels subjected to subzero temperature for a determined period. Reproduced from ref. [145] with permission from Elsevier, copyright 2021.

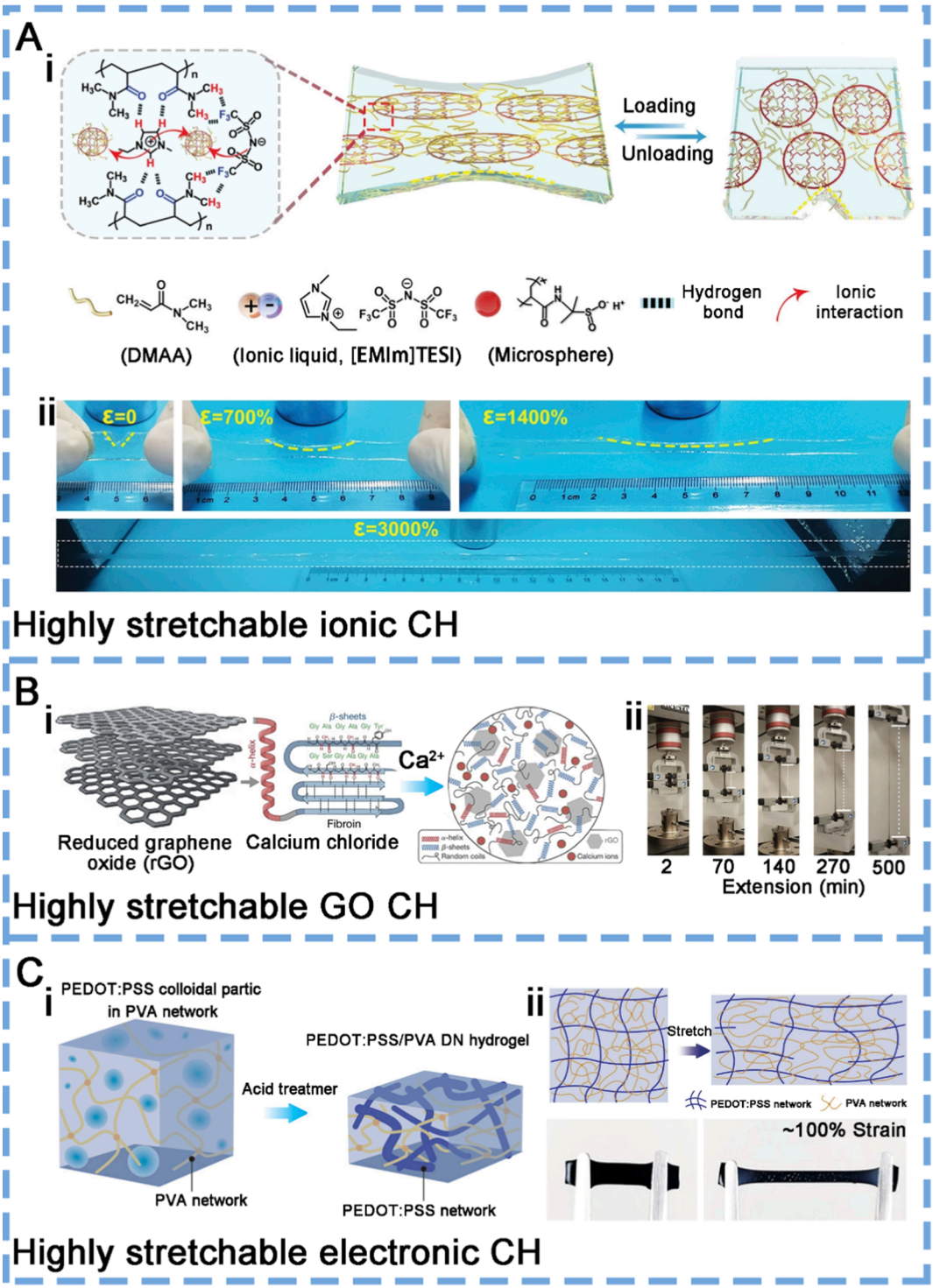

Fig. 5. Representative methods to improve stretchability. (**A**) Stretchable ionic CHs. i) Schematic illustration of ionogels. iii) Images of the stretch process from 0 to 3000%. Reproduced from ref. [166] with permission from Wiley-VCH, copyright 2022. (**B**) Highly stretchable GO CHs. i) The components used for preparation of this CHs. ii) Photographic images showing tensile stretching of CHs with an Instron mechanical tester. Reproduced from ref. [15] with permission from Wiley-VCH, copyright 2022. (**C**) Stretchable electronic CHs. i) Schematic illustration of the fabrication and structural change of PEDOT:PSS and PVA acid treatment process. ii) Schematic illustration of the deformation mechanism of the PEDOT:PSS/PVA DN hydrogel and images of DN hydrogel under stretching. Reproduced from ref. [169] with permission from Wiley-VCH, copyright 2022.

3.2. Water retention and stretchability

As the largest part of CHs, water content dominates the main properties of CHs, including self-healing, adhesion, electrical performance, and particularly stretchability [46]. On the other side, the ability of CHs to maintain their water content is also an important indication of their environmental adaptability. Water retention for CHs bioelectronics means the limitation of water evaporation in dry conditions and the anti-freezing abilities [135,136] once the temperature decreases below the freezing point of water [137]. Conventional hydrogels which use pure water as a dispersion medium will be unsoftened at freezing environments, severely limit their wide applications [138]. Inspired by freezing tolerance of plants, the lipid species in cell membrane has been employed to stabilize the hydrogels during freezing [139,140]. Recently, organic solvent substitutions have been used to synthesize hybrid organo-CHs that maintain stable flexibility and stretchability at dry and freezing (<-20 ℃) environments (Fig. 4A) [135,140,141]. However, the use of organic liquids such as propylene and ethylene glycol as solvents for water within CHs cannot ignore their environmental impacts and biotoxicity. As an alternative, Dimethyl sulfoxide (DMSO) [96,142] and glycerol [143,144] with good biocompatibility have been used for inhibiting the freezing and volatilization of CHs. For ionic CHs, their conductivity will decrease due to the reduced ion dissociation in organic solvent and the increased viscosity of the solution, affecting ion mobility. As a solution for this challenge, water is used to compose binary solvent systems for CHs [96].

The simplest and most effective way to composite CHs with good water retention is introducing serval salts (*e.g.,* Li^+ [138,145], Na^+ [96], Ca^{2+}, Zn^{2+} [143,146], Al^{3+} [147]) or polyelectrolytes [148] into the hydrogel matrix. For example, $CaCl_2$ has been used to prevent roads from icing over [149], by mixing the salts into CHs precursor solutions or immersing the prepared hydrogels in the salt solutions for ions diffuse into CHs (Fig. 4Bi) [145,150]. Introducing salts and polyelectrolytes can increase their ionic conductivity for CHs by increasing free-moving ions [151]. Colligative property of ionic compounds can also depress the freezing point of aqueous phase for anti-freezing CHs (Fig. 4Biii). However, for some ionic CHs, water evaporation at the surface still exists. The presence of water molecules within the hydrogel network can increase the ionic mobility of CHs, improving its conductivity. Methods like Binary solvents CHs or salts such as LiCl are also used. LiCl can trap water molecules at low relative humidity (RH) environments (<30% RH) [152]. LiCl-CHs can bind the water to reduce the vapor pressure of CHs and achieve water re-generation after vacuum drying (Fig. 4Bii). On the other hand, compared with pristine CHs, incorporating different ions and organic liquids into hydrogel matrixes will enhance their crosslinking density, which is beneficial for their stretchability. Nevertheless, it is also noticed that denser hydrogel networks suppress ion transporting and reduce ionic conductivity. Achieving a balance between them is equally important. Ionic CHs based on ionic liquid have gained great attention due to their nature of non-volatility, stability, and ionic conductivity [153,154]. Whereas the cost and the inevitable biotoxicity of ionic liquids are not suitable for applications in biological tissue.

Though water retention can maintain CHs properties in harsh environments, most of them will over-expand in aqueous environment due to their rich hydrophilic groups, leading to deformation and unstable performance [155]. Serval methods such as solvent replacement and metal ions doping which can increase the cross-linking density and adjust the polymer-water interaction have been utilized to tackle this issue [156,157]. Water also plays a significant impact on stretchability for CHs. The water content within CHs network acts as lubricant, reducing the forces required for deformation and enabling the CHs to stretch more easily. This effect is particularly pronounced in hydrogels with high water content [158–160]. Moreover, water retention affects the mechanical properties of the hydrogel. The presence of water molecules within the hydrogel network increases its overall swelling capacity, making the hydrogel softer and more compliant. This increased compliance enables the hydrogel to stretch further before experiencing failure, leading to increased stretchability [161,162]. For CHs-based devices, CHs with good water retention ability can maintain the properties (conductivity, stretchability, self-healing, *etc.*) for long-term operation. Furthermore, CHs that can capture moisture from environment could provide continuous water resource for hydrovoltaic devices.

Stretchability is crucial demand for CHs-based bio-applications. Methods like increasing crosslinking density by adding different crosslinkers or/and ions to synthesize stabler polymer networks or/and dual network within CHs for energy dissipation during stretching has been used for different types of stretchable CHs [151,163–165]. Besides, adding retractive microspheres (Fig. 5Ai) or silk-based materials (Fig. 5Bi) into hydrogel matrix can enhance their stretchability. For example, a highly stretchable (>3000%), recyclable and repairable ionic CH with suitable ionic conductivity (1.34 S/m) has been composited by introducing microsphere into hydrogel matrix (Fig. 5Aii) [166]. For ionic liquid CHs, an ultrastretchable ionogel (10250%) has been rationally designed through water-induced competitive hydration interactions [167]. Furthermore, different types of nanomaterials (CNT [126], GO [85], MXene [12,146], *etc.*) are modified with hydrophilic substance, oxidised to increase their dispersibility, or loaded with hydrophilic nanomaterials to improve their additivity for CHs matrix and inhibit phase separation (Fig. 5Bi). Large number of hydrophilic groups can be well cross-linked to the polymer chains or/and stretchable materials to enhance CHs stretchability. Along these lines, multifunctional CHs have been synthesised by mixing silk fibroin and reduced graphene oxide (rGO). Hierarchal bonding scheme between rGO and polymer chains gives rise to high stretchability with a the record-breaking elongation capacity of ~25000% (Fig. 5Bii) [15].

It is also a great challenge to achieve high conductivity (>10 S/cm) and good stretchability (>100%) simultaneously. Especially for conductive polymer-based CHs whose conductive networks are brittle due to the low dissipate strain energy ability for these materials (strain at break < 100%) [7,158,163]. Incorporating another stretch network to protect conductive networks during stretch process can enhance their stretchability, but with a huge decrease in conductivity. Such a phenomenon is attributed to the introduction of electrically insulating network, the continuity of the conductive network deteriorates and the content of conductive materials is low (<1 wt%) [168]. For conductive polymer-based CHs, PEDOT:PSS is the most promising conducting polymer. Pure PEDOT:PSS hydrogels have been reported to present a high conductivity (40 S/cm), while its stretchability is much lower than normal bioelectronics [10]. The state-of-the-art methods to composite high performance PEDOT:PSS-based CHs include acid treatment [169] and bi-continuous phases [170]. These CHs with high PEDOT:PSS concentration (>5 wt%) can provide much higher electrical conductivity

(>10 S/cm) and suitable strain (>150%) for bioelectronic applications. For acid treatment, acid replace the water of as-made CHs. After acid treatment, the colloidal particles of PEDOT:PSS are aggregated within the PVA network. Therefore, PVA network acts as a template to constrain the aggregation of PEDOT:PSS into a continuous conductive network (Fig. 5Ci). After first stretch loading, PEDOT:PSS network breaks and forms a network with lager dispersed pocket size without affecting the continuity of electronic transmission. This partially ruptured network is more stretchable, and does not rupture further in the following stretch due to the significant energy dissipation of the PVA network (Fig. 5Cii). As for bi-continuous PEDOT:PSS-CH, the continuous conductive network has been wrapped by stretchable polyurethane, achieving a slight trade-off between mechanical performance and conductivity for CHs.

3.3. Self-healing ability

Biological systems can self-heal. In contrast, electronic devices degrade over time due to fatigue, corrosion or damage during operation, leading to device failure [171,172]. For CHs-based electronics, self-healing is crucial to remain their original properties, such as mechanical, conductive and rheological abilities in a short time after unpredictable damage [173–175]. Autonomous self-healing is a process that a material can heal damage intrinsically and automatically, restoring itself to normality [173]. There are two major approaches for self-healing CHs systems, including dynamic covalent reactions (chemical crosslinking) and noncovalent reactions (Fig. 6A) [176]. Some self-healing CHs depend on external stimulus such as heating and UV illumination [177], whereas others rely on the autonomous interactions [178]. The advantages of autonomous self-healing have aroused much interest in preparing CHs for bioelectronics. For CHs-based self-powered sensing devices, good self-healing CHs can recover from cracks and restore their mechanical and electrical properties after numerous operations. Such ability that provided by CHs allows CHs-based self-powered devices to recover their critical functionality from damage, enabling durable and highly sustainable device operations.

Covalent interaction for CHs self-healing can be classified as ion chelation and dynamic covalent bonds. Chelation interaction mainly relies on several coordinate bonds between ligands (polymers) and positively charged transition-metal ions [179]. The transition-metal ions are surrounded by the ligands which donate electrons with these ions. Meanwhile, due to the lattice structure of metal complexes and the many donor atoms involved, this interaction is more intense than covalent bonds. The chelation interaction

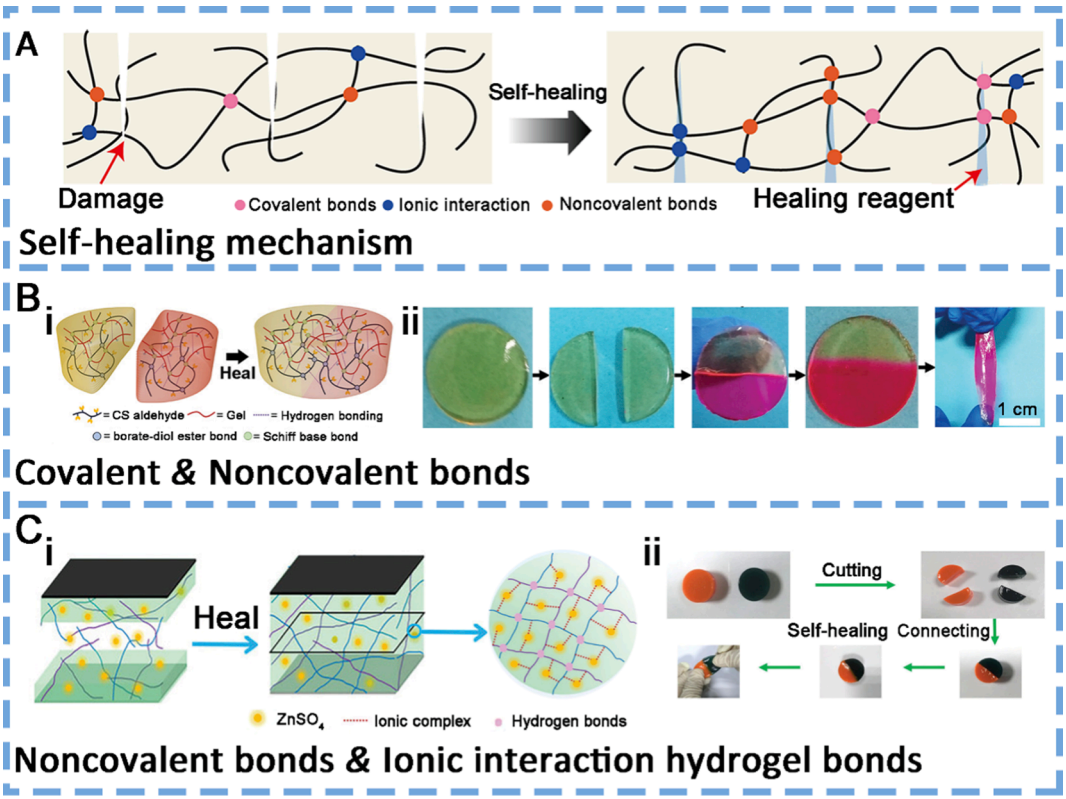

Fig. 6. Self-healing ability. (**A**) Schematic illustration for self-healing of CHs. (**B**) Multiple interactions for self-healing. i) Schematic of the self-healing mechanism (hydrogen bond, borate-diol ester bond, and Schiff base bond) and ii) images showing the macroscopic self-healing behaviour of CHs. Reproduced from ref. [184] with permission from Wiley-VCH, copyright 2021. (**C**) Noncovalent bonds & ionic interaction for CHs self-healing. i) Mechanism and ii) Photographs of the self-healing process for $ZnSO_4$/sodium alginate/polymethylacrylic acid (SPMA-Zn_3) hydrogel. Reproduced from ref. [190] with permission from Wiley-VCH, copyright 2022.

can display high adhesion and reversibility at the same time. Natural mussels form sticky and elastic feet through chelation between Fe^{3+} and catechol. This unique property has inspired the synthetic of various self-healing CHs [141,176,180–182].

Various self-healing CHs have been produced with dynamic covalent bonds, of which imine bonds are the most widely used. Schiff-based reaction is the interaction between amine and aldehyde groups, forming imine bonds [176,183,184]. Another dynamic covalent bond is diol-borate ester bond, which can be reversibly broken and formed under physiological conditions, depending on the reactions of diols and boronic acid. For example, PVA with diol groups is frequently used to prepare diol-borate ester bond-based CHs (Fig. 6Bi-ii) [185,186]. Other types of dynamic covalent, including Diels-Alder and acyl hydrazone bonds, are less used for CHs due to the need for external stimulation, such as heating and acidic environment to accelerate self-healing reactions [187,188].

Noncovalent interaction for self-healing CHs including hydrogen bond [67,189], ionic interactions [190], π–π stacking interactions, host–guest interactions [191], and hydrophobic interactions [110]. Hydrogen bonds are the most common noncovalent bonds in nature, which can form between positive hydrogen atoms and electronegative acceptor atoms such as oxygen, nitrogen, or fluoride. Large amount of hydrogen bonds within CHs matrix can significantly improve the self-healing properties, stretchability, and adhesion, which are highly desirable for a wide range of applications [192]. However, among both covalent and noncovalent bonds, hydrogen bonds exhibit the weakest bonding strength, ranging from 0.25 to 15 kcal/mol [193].

However, the inconsistency between CHs toughness and rapid self-healing remains unaddressed, still facing grave challenges in the dynamic and mechanically demanding environment of human tissues. Although some CHs seem to achieve rapid self-healing with good efficiency, these properties come at the cost of the weakness of elasticity and mechanical strength [176]. Existing self-healing CHs mainly depend on noncovalent bonds that exhibits quick recovery ability. A rapid self-healing CH has been prepared by grafting citric acid and ascorbic acid. Dynamic borax bonds and hydrogen bonds significantly improve the self-healing properties of CHs, achieving a stress self-healing efficiency 99.56% within 2 min [194].

Ionic interactions occur when the metal ions link with their ligands, relying on the electrostatic interactions between oppositely charged moieties (Fig. 6Ci-ii). On the other hand, host–guest interactions refer to the selective accommodation of guest molecules in subject molecules, such as macrocyclic molecules, which form inclusion complexes through π-π stacking (which occurs between aromatic rings) and hydrophobic interactions [176]. Different bonds cooperate together by forming CHs with excellent self-healing efficiency (>95%) [190]. For example, CHs with excellent self-healing efficiency (97%) have been synthesized through dynamic cross-linking bonds, including hydrogen bonds and π–π stacking [195].

To date, the easiest and most effective way for hydrogel systems with both self-healing and good conductivity is adding nanomaterials with hydrophilic groups such as MXene [196,197], GO [198], and carbon nanotubes [199], which can achieve nanoreinforcement by dynamic interactions between polymer groups.

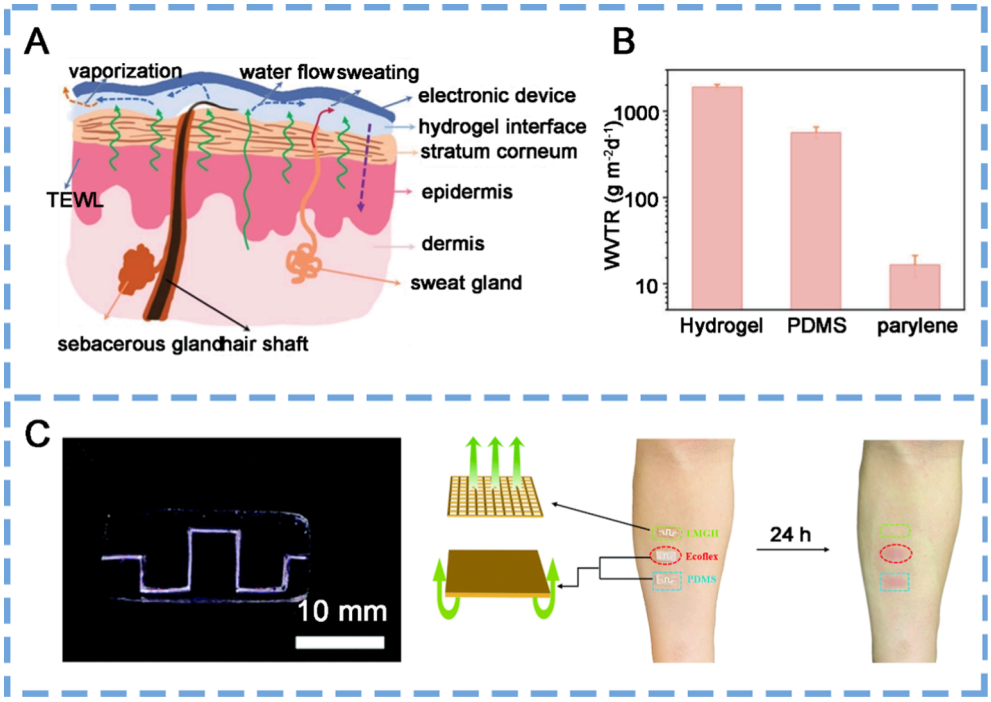

Fig. 7. Air-permeability. (**A**) Schematic illustrations for mechanisms of air-permeable adhesion with human skin. (**B**) The comparison of water vapor transmission rate for a 50 μm-thick hydrogel film containing 50% glycerol, a 50 μm-thick PDMS, and a 3 μm-thick parylene film. Reproduced from ref. [203] with permission from Wiley-VCH, copyright 2023. (**C**) Images showing the skin irritation results of different materials. Reproduced from ref. [106] with permission from Royal society of chemistry, copyright 2022.

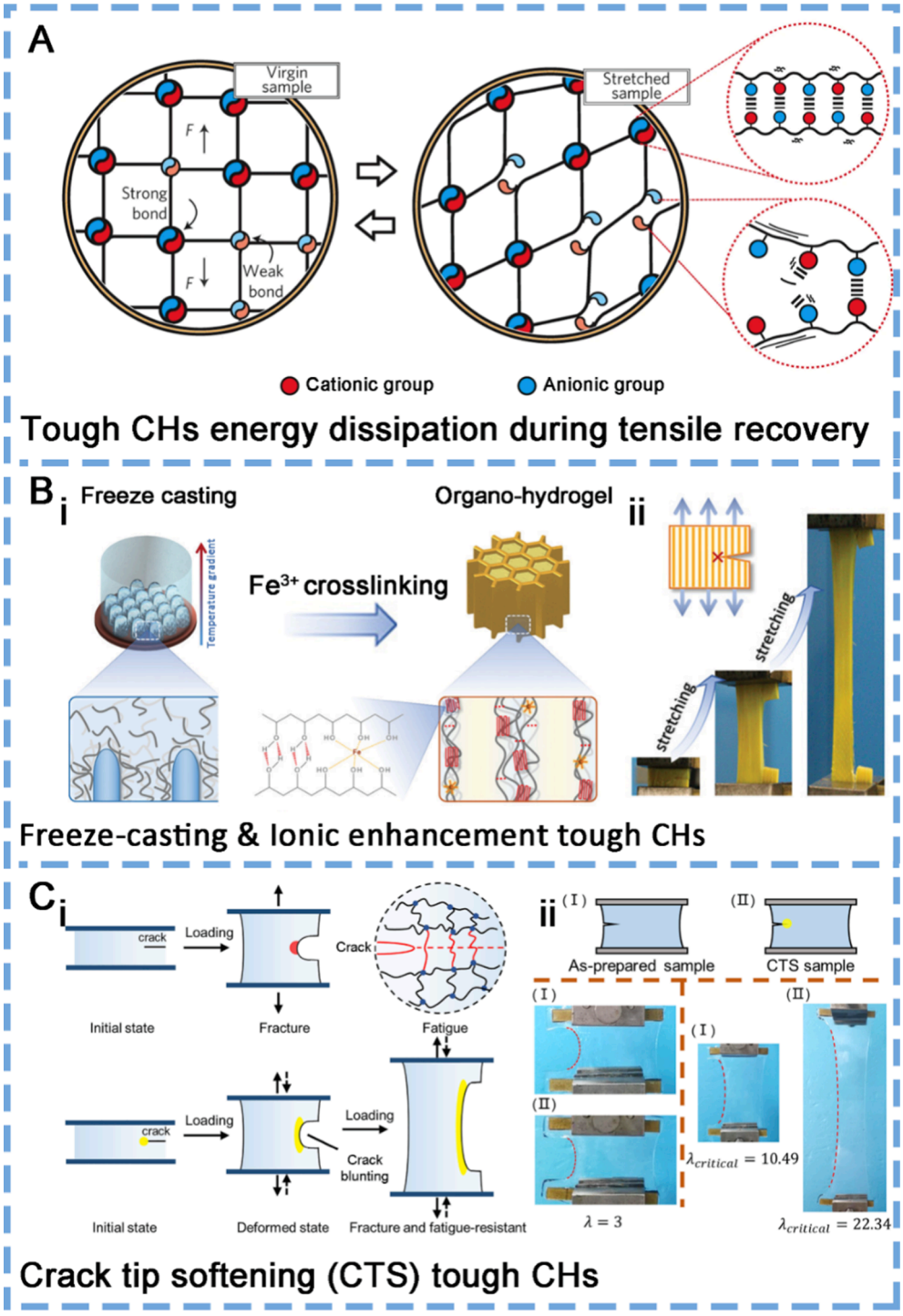

Fig. 8. Toughness and state-of-the-art methods to composite tough CHs. (**A**) Illustration of polymer networks with different strength. Reproduced from ref. [221] with permission from Springer Nature Limited, copyright 2013. (**B**) i) Schematic diagrams of freeze-casting assisted solution substitution strategy of the PVA organoCH. ii) Images of a sample being stretched. Reproduced from ref. [226] with permission from Wiley-VCH, copyright 2022. (**C**) i) Schematic illustration of crack tip softening makes the hydrogel crack blunt and resists the crack propagation under both monotonic load and cyclic load. ii) Images of crack blunting in the as-prepared and CTS hydrogel samples. Reproduced from ref. [230] with permission from Proceedings of the National Academy of Sciences, copyright 2023.

3.4. Air-permeability

For long-term health monitoring, air permeability is another crucial issue for wearing comfort and preventing on-tissue inflammation [200,201]. Air permeability for on-skin wearable electronics relies on the degree of breathability that allows free water vapour and oxygen exchange between the human skin and the external environment (Fig. 7A). The water vapor transmission rate (WVTR) of hydrogels are characterized in comparison with PDMS and parylene (Fig. 7B) and reaction applied to the skin for 24 h (Fig. 7C) [202]. Due to the large amount of water and the inherent porosity within hydrogels, CHs-based wearable electronics have a higher water vapour transmission rate than human skin. External oxygen can exchange gas with water vapour from internal exudate, allowing virtually unimpeded *trans*-epidermal water loss and free breathing under the tissue [203]. As substrates, hydrogels based-nanofibrous materials (such as metal-nanowires [204], and PEDOT film [11]), meshes, and ultrathin skin-electronics are also profitable strategies to fabricate bioelectronics with full integration of high sensitivity, high stretchability, as well as outstanding breathability [106,205,206]. For example, an ultrathin hydrogel-interfaced flexible (opto) electronics without causing skin irritation for over one week has been demonstrated [203]. The skin-similar air permeability of CHs will promote the general applicability of tissue-integrated electronics that act not only as substrates but also as conductive devices for bioenergy harvesting and self-powered sensing.

3.5. Toughness

Compared with stretchable elastomers, conventional hydrogels are brittle with a single network and suffer from low mechanical strength and toughness. Single networks are commonly toughened by introducing sacrificial bonds, but during the stretch-resilience process, these bonds break and possibly reform, causing pronounced hysteresis [195,207–212]. These hydrogels show a low toughness between 10 and 100 J/m^2, restricting their wide applications in various environments [42]. Typically, CHs with high tensile stress (>0.1 MPa) and fracture energy (>100 J/m^2) can be considered as tough CHs [213,214]. The toughness of hydrogels has been attributed to the combination of two mechanisms: crack bridging by the covalent crosslinks and hysteresis by unzipping the network of ionic crosslinks. To improve the toughness of CHs, the general principle is introducing tougher network into CHs system to dissipate mechanical energy under large deformation and maintain its original characteristics after deformation (Fig. 8A). Methods focusing on the reconstruction and enhancement of hydrogel networks have been used recently, such as establishing DNs [215], topological networks [216], anisotropic structures [217], slip ring structures [218], hydrophobic associations [219], and ionic enhancement [220].

To date, toughening strategies like DN and ionic enhancement have been widely used. DN hydrogels consist of a stiff brittle, and a soft extensible network. The breakage of a stiff brittle network near the crack tip will dissipate gel deformation energy, while soft extensible network will maintain the macroscopic integrity of the hydrogels [221–223]. A type of DN ionic hydrogel for electronic skin

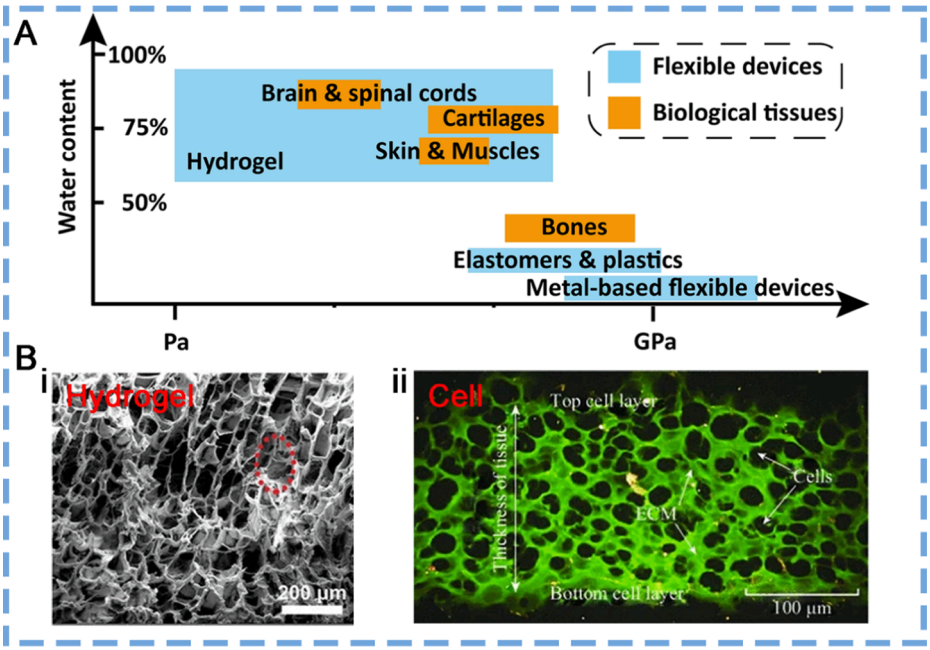

Fig. 9. Common flexible devices Young's modulus, water content, and representative structure of CHs compare with Human tissues. (**A**) Current flexible devices Young's Modulus and water content compare with biological tissues. (**B**) i) Morphology image of PSGO-PEDOT-PAM CHs. Reproduced from ref. [73] with permission from Wiley-VCH, copyright 2020. ii) A cross-section of a multicellular layer of tissue (Areas marked in green belong to extracellular matrix, while the unstained regions represent cells). Reproduced from ref. [264] with permission from Springer Link, copyright 2015.

has been designed [224]. Attributing to the combination of physically crosslinked agarose and covalently crosslinked networks, this CH has good mechanical performance, with a stretchability of 400% and toughness of 1826 J/m^2. On the type of ionic enhancement, CHs can be toughened by mixing weak and strong bonds, such as ionic crosslinks with polymer chains and hydrogen bonds between polymer chains. During the stretch process, energy is dispersed and dissipated, and the stretchable networks (covalent bonds) remain intact and stabilize the deformation. As the material is stretched, the ionic crosslink bonds gradually unzip starting with those closest together, and continuing with more widely spaced crosslinks as the stretch increases [220,225]. A super-tough organo-CH has been proposed through freeze-casting assisted solution substitution [226]. This strategy enables the formation of CHs in one step with exquisite hierarchical anisotropic structures coupled with synergistic strengthening and toughening effects across multiple length scales (Fig. 8Bi). This CH exhibits high strength (6.5 MPa), high stretchability (1710% in strain), ultra-high toughness (58.9 MJ/m^3) (Fig. 8Bii). For some other state-of-the-art methods, ionic CHs through water-induced hydration interactions have been designed, which are deliberated and controlled within ionic liquid-based hydrogels to achieve a comprehensive performance (high stretchability of 10250%, high toughness of 21.8 MJ/m^3, and ionic conductivity of 0.7 S/m) with high environmental stability for flexible electronics [167].

CHs with high toughness have greatly expanded the range of real-world applications for CHs-based devices. Designed CHs structure with excellent mechanical performance also make great strides. PVA-based CHs can be toughened by repetitive freezing-thawing processes attributed to the directional arrangement of ice crystals, building an orderly network structure [227–229]. Recently, a concept of “crack tip softening” has been proposed to simultaneously improve the fracture toughness and fatigue threshold of single polymeric network (Fig. 8Ci) [230]. This strategy can soften the crack tip portion by external field stimulation. In this way, the softened crack tip is blunted under monotonic or cyclic loading, relieving the stress concentration. In addition, the softened crack tip has better elasticity and can provide an elastic shield for the bulk phase material, effectively resisting crack extension (Fig. 8Cii).

3.6. Biocompatibility

Hydrogels are a promising class of materials for biomedical applications. CHs-based electronics can be implanted into human body for tissue engineering [231,232], drug delivery [233,234], and sensing [235] due to their excellent biocompatibility [236–238]. These materials have been shown to exhibit low cytotoxicity and minimal cellular response, making them ideal for use in various biomedical applications. CHs with porous networks can replicate the mechanical and physical properties found in natural tissues, such as elasticity, stiffness, and permeability [239–241]. Furthermore, their high-water content, which can reach up to 95%, enhances their biocompatibility as it mimics the aqueous environment of living tissues (Fig. 9A) [242–244]. Unlike other conductive materials, such as liquid metals, which contain large amounts of toxic metal ions that can lead to oxidative stress and cellular damage, the material being discussed here poses a lower risk to cells [245]. Additionally, these materials have mechanical properties that are vastly different from those of biological tissues, which can negatively impact the growth and proliferation of cells [246].

The structure of CHs is another factor that contributes to their biocompatibility. Hydrogels can be formed from a wide range of polymers, including natural polymers such as collagen [247] and gelatin [248], and synthetic polymers like polyvinyl alcohol [231] and polyethylene glycol [198]. The cross-linking of these polymers can be achieved through physical or chemical methods, which allows for forming hydrogels with tunable mechanical and physical properties. The structure of CHs is highly porous, allowing for the transport of nutrients and waste products, which is essential for cell survival (Fig. 9Bi-ii). In terms of their mechanical properties, CHs have been shown to have Young’s modulus similar to that of human tissue. This is important as it allows the hydrogel to support the growth and proliferation of cells while preventing excessive deformation. The high-water content of hydrogels also contributes to their mechanical properties, as it allows the hydrogel to exhibit a high level of viscoelasticity, similar to that of biological tissues (Fig. 9A) [249–253].

When foreign materials or devices are introduced into human body, foreign body response (FBR) is also a key consideration. For CHs-based bioelectronics, which involves the integration of electronic components with biological systems, an immune-mediated reaction that can influence its functionality and long-term performance of these devices. One of the primary concerns with the FBR is the formation of a fibrous capsule around the implanted device. This capsule, composed mainly of collagen fibers, can isolate the device from the surrounding tissue and hinder its functionality. Moreover, the capsule may limit the delivery of oxygen and nutrients to the device, leading to its degradation over time [254–256].

To mitigate the foreign response and improve the biocompatibility of CHs-based bioelectronics, various strategies are being explored. Surface modifications of the device materials can be employed to make them more compatible with the surrounding tissue and reduce the immune response [257]. Coating the device surface with bioactive molecules, such as cell adhesion peptides or anti-inflammatory agents, can promote cell-material interactions and reduce the immune response [258,259]. Additionally, the controlled release of immunomodulatory agents from conductive hydrogel-based bioelectronics represents a promising approach to modulate the FBR [260,261]. Localized delivery of anti-inflammatory drugs or growth factors within the hydrogel matrix can attenuate inflammation, promote tissue healing, and mitigate fibrous encapsulation [262,263]. These strategies aim to create a favorable microenvironment for device integration and long-term functionality.

The FBR remains a critical consideration in the development of conductive hydrogel-based bioelectronics. Understanding the interactions between conductive materials, hydrogel matrices, and host tissues is essential for optimizing biocompatibility and long-term performance. Surface modifications and incorporation of immunomodulatory agents represent promising strategies to mitigate the FBR, enhance tissue integration, and improve the overall biocompatibility of conductive hydrogel-based bioelectronic devices.

3.7. Fatigue stability

Conventional CHs are limited by their poor fatigue stability, which impede their long-term performance in practical applications. Fatigue stability refers to the ability of a material to maintain its performance over time, even after undergoing repeated cycles of deformation or stress. In the case of CHs, fatigue can lead to the formation of cracks, voids, and other defects that can disrupt the conductive network, which will decrease the electrical conductivity and mechanical strength [265–267]. Furthermore, the migration of conductive particles can also contribute to the decrease of conductivity over time. In order to overcome these limitations and enhance the long-term performance of CHs, a range of methods have developed to improve their fatigue stability.

One approach to improving the fatigue stability of CHs is introducing nanocomposites into CHs systems, such as MXene, carbon nanotubes, nanoclay, and metal-based nanoparticles. The use of these nanoparticles has been shown to reinforce the CHs matrix and improve their mechanical and electrical properties, as well as the fatigue stability [268,269]. MXene nanosheets composited into hydrogels matrix (Fig. 10Ai) has been proposed. Hydrophilic MXene formed strong interactions with the polymer matrix and endowed the CHs with excellent mechanical strength, long-lasting stability and fatigue resistance, and conductivity (Fig. 10Aii) [269].

Another approach to improve the fatigue stability of CHs is designing crosslinked structures. Crosslinking refers to forming chemical bonds between different parts of the material, creating a more stable network that can withstand repeated deformation cycles. Crosslinked CHs have been shown to exhibit improved fatigue stability compared to non-crosslinked hydrogels, as the crosslinked structure helps to prevent the migration of conductive particles and the formation of cracks and voids [271–273]. Recently, a highly stretchable and elastic ionic-CH has been designed. This nanoclay based CH exhibited excellent tensile, fatigue properties and conductivity due to the structural-mechanical–electrical integrity of the nanoclay crosslinked and nano-reinforced interpenetrating network [274].

In addition to the use of nanocomposites and crosslinked structures, other methods have been developed to improve the fatigue stability of CHs, including the use of mechanical reinforcement, such as fibrous structures, and the use of different crosslinking agents,

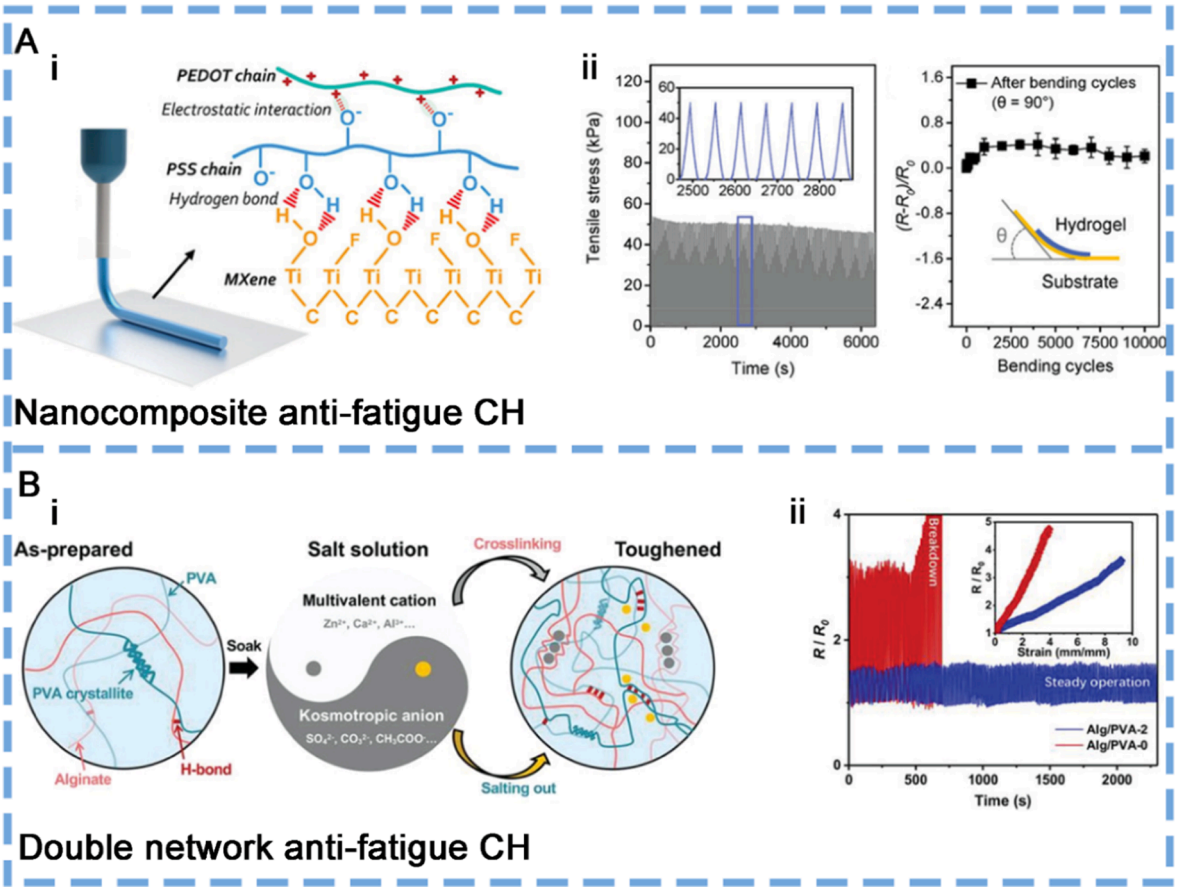

Fig. 10. Representative sample to composite CHs with good fatigue stability. (**A**) Nanocomposite anti-fatigue CH i) Scheme illustrating the preparation of MXene-functionalized PEDOT:PSS ink for extrusion printing. ii) The tensile stress–strain curves of the printed hydrogel at a strain of 5% for>100 cycles and electrical resistance variations as a function of bending cycles (90°). Reproduced from ref. [13] with permission from Wiley-VCH, copyright 2022. (**B**) Ionic reinforcement of anti-fatigue CH. i) Schematic diagram of the synthesis of PVA ion-reinforced CHs. ii) Cyclic performance of the notched sensor at a strain of 200%. Reproduced from ref. [270] with permission from Wiley-VCH, copyright 2022.

adding reversible crosslinked networks for CHs (Fig. 10Bi) [275–278]. Despite these advances, there is still much to be done to fully understand the mechanisms of fatigue degradation in CHs and to develop new and more effective methods for improving their fatigue stability (Fig. 10Bii). In addition, improved fatigue stability is also critical for their long-term use in bioenergy harvesting and self-powered applications.

The development of CHs with comprehensive properties in interfacial adhesion with various surfaces, good water retention and stretchability to meet the needs of different environments, quick self-healing ability, good air-permeability and biocompatibility, and high toughness and fatigue stability is still a significant challenge. For improving various abilities for long-term bioenergy harvesting and self-powered applications, further research is needed to fully realize these materials' potential and enhance their performance in practical applications.

4. Bioenergy harvesting devices

CHs with comprehensive properties can satisfy long-term, precise, and continuous operation. However, conventional CHs-based electronics rely on external power supplies and complex related fabrications for ecosystems running, limiting their portable applications [279–281]. Converting bioenergy to electrical energy in human daily life as sustainable power sources for wearable devices has gained considerable attention [279,282]. In this section, we will introduce various CHs-based devices for bioenergy harvesting, mainly focusing on triboelectrics, piezoelectrics, hydrovoltaics, thermoelectrics, and biofuels. Comparisons of CHs-based devices on material functions, mechanical performance, and device duration have been conducted and summarized inTable 3.

4.1. Triboelectric nanogenerators (TENGs)

TENG is a promising technology for harvesting mechanical energy due to the easy device fabrication, ability to harvest a wide range of mechanical energies, and diverse material options [283–287]. TENGs are based on a combination of contact electrification and

Table.3
Comparative performance and mechanical properties of wearable CHs-based bioenergy harvesting devices.

Typical materials	Device type	CH functions	Max strain	Time	Ref.
PAM/LiCl	TENGs	Electrodes	1160%	Long-term	[301]
PAM/LiCl	TENGs	Electrodes	2000%	Long-term	[298]
PVA	TENGs	Electrodes	35%	Long-term	[302]
PAM/NaCl	TENGs	Electrodes	/	Long-term	[382]
PAM/PEDOT:PSS	TENGs	Electrodes	2850% (Hydrogel) 300% (TENG)	Long-term	[383]
PVA/MXENE	TENGs	Electrodes	200% (Hydrogel)	Long-term	[304]
PAM/NaCl	TENGs	Electrodes	/	Long-term	[306]
PAM/SA	TENGs	Electrodes	740% (Hydrogel)	Long-term	[43]
PGA/PEDOT:PSS	TENGs	Electrodes	650% (Hydrogel)	Long-term	[67]
PAM/PDA	TENGs	Electrodes	6000% (Hydrogel)	Long-term	[384]
PAM/BTO NCs	TENGs	Electrodes	800% (Hydrogel) 433.3% (TENG)	Long-term	[303]
CPPH	TENGs	Electrodes	1530%	Long-term	[305]
PAM/CNTs	TENGs	Electrodes	4196% (Hydrogel) 500% (TENG)	Long-term	[45]
PHEMA-GO	PENGs	Substrate	98.5% stress	Long-term	[321]
BC/$ImClO_4$	PENGs	Substrate	17% stress	Long-term	[322]
PVA/PVDF	PENGs	Substrate	300.8% stretch	Long-term	[325]
PAAM/NaCl	Piezonic	Piezonic	/	Long-term	[328]
PAN/PVDF	PENGs	Substrate	90% stress 175% stretch	Long-term	[327]
CHACC/PEDOT:PSS/PVDF-TrFE	PENGs	Substrate	55% stress 293% stretch	Long-term	[324]
IPHCs	MEGs	Water resource @protons path	/	Long-term	[40]
PVA/FCB/3DS	EIGs	Water resource	120° bending strain	Over 150 h	[342]
Gelation/Al_2O_3	EIGs	Heat conduction	180° bending strain	Over 1000 h	[344]
PVA-PA	MEGs	MEG	2.2 cm bending radius	Long-term	[347]
AA/GO	TEGs	Cooler	/	Long-term	[359]
/	TEGs	Cooler	2 cm bending radius	24 h	[360]
PEDOT:PSS	TEGs	p-type	30% (Hydrogel)	Long-term	[41]
BC/IL	TEGs	Substrate	40.99%	Long-term	[361]
PVDF-HFP	TEGs	Substrate	/	Long-term	[385]
PVD-HFP/EMIM:DCA	TEGs	Substrate	/	Long-term	[364]
ASD HEMA	BFs	Substrate	180° bending strain	2.5 h	[374]
DMA	BFs	Glucose carriers	/	/	[386]
PAAM	BFs	Glucose carriers	60%	/	[375]

Sodium alginate (SA), $BaTiO_3$ nanocubes (BTO NCs), 2, 2, 6, 6-tetramethylpiperidine-1-oxylradical-oxidized CNFs/ polyaniline-PVA/borax (CPPH), Carbon nanotubes (CNTs), Polyhydroxyethyl methacrylate (PHEMA), Graphene oxide (GO), Bacterial cellulose (BC), Imidazolium perchlorate ($ImClO_4$), Polyvinylidene fluoride (PVDF), Poly acrylamide (PAAM), Polyacrylonitrile (PAN), Chitosan quaternary ammonium salt (CHACC), Poly (vinylidene fluoride-co-trifluoroethylene) (PVDF-TrFE), Ionic polymer-hydrogel-carbon composites (IPHCs), Functionalized conductive carbon black (FCB), Three-dimensional sponge (3DS), Phytic acid (PA), Moisture-electric generator (MEG), Acrylic acid (AAc), Bacterial cellulose (BC), Ionic liquids (ILs), Poly(vinylidene fluoride-co-hexafluoropropylene) (PVDF-HFP), 1-ethyl-3-methylimidazolium dicyanamide (EMIM:DCA), 2-hydroxyethyl methacrylate (HEMA), Dopamine methacrylamide (DMA).

electrostatic induction. When two materials come into contact, electrification occurs to produce net static charges with opposite signs in the surfaces of the two materials. This electrification process is correlated to the electron transfer between two materials with different electronegativity or tribonegativity, that the one with higher electronegativity tends to capture electrons from the lower. The relative motions of the electrified surfaces then lead to the time-variation of the electrostatic fields of the generated static charges, which further produces the displacement current inside the TENG and induces conduction current through the external circuit [288,289]. As mechanoelectric energy converters, TENGs are increasingly being incorporated with flexible conductive materials for bioenergy harvesting [290], self-powered sensing [291], and human–machine interaction [292]. In many application scenarios, non-toxicity, softness, stretchability, transparency or biocompatibility is typically required. CHs have then emerged among the top candidate materials for such devices [293–296]. CHs are employed as conductive electrodes in TENGs as an ionic electrode to replace conventionally used rigid non-stretchable metal electrodes. As opposed to other flexible electrodes (carbon nanotubes, graphene, silver nanowires, *etc.*), hydrogels with higher stretchability dominate TENGs. The other advantage of CHs-based TENGs is their resistance changes during the deformation process are much smaller than these electronic conductors, which is more assessable for mechanical energy harvesting [297]. To date, almost all types of CHs have been attempted as flexible and stretchable electrodes inner CHs-based TENGs for charge collection. Incorporating different flexible dielectric tribo-electrification materials (PDMS [298], PTFE [299], and silicone rubber [300], *etc.*), various CHs-based TENGs have been composed, adopting representative TENG structures including single-electrode mode [301], contact-separation mode [302], and sliding modes [301]. By selecting appropriate tribo-materials and TENG structures, the above discussed advantages of CHs materials can be maintained in the final TENG-based devices.

The single-electrode mode TENG is the most commonly employed structure for CHs-based TENGs, in which CHs are designed as back electrodes and attached to a dielectric soft electrification layer (PDMS, Ecoflex, *etc.*). The electricity generation mechanism is about the same as the conventional TENG using electronic electrode, but the difference is as following: with an electronic conductor electrode, the induced conduction current flows directly through the back electrodes to balance the static charges of the electrified dielectric layers; with the CHs ionic electrode, ions in the gel electrolyte are attracted to screen the unbalanced static charges, and meanwhile, the electric double layer (EDL) formed between the gel and the external circuit exchanges the ionic flux in the gel and the electron flux in the external circuit (Fig. 11A). It is assumed that there is no Faradic reaction inside the EDL capacitor, and which is also supported by the characteristics of TENGs, i.e. the output alternating current and low charge quantity. As for the single-electrode mode TENGs, the contacting materials can be various, such as human skin [67], gloves [45], metal layers [302], *etc.* This type of CHs-based energy harvester has been most widely used for mechanical energy harvesting in human daily activities, such as hand-tapping, finger/knee-bending, and footsteps (Fig. 11B) [303]. Due to the high stretchability, flexibility, and transparency of the CHs, CHs-based TENGs are flexible enough to withstand various deformation including knotting, twisting, and bending, and have been demonstrated for applications in electronic skins and soft robots. (Fig. 11C) [301]. Utilizing the PAAm-LiCl hydrogel as the electrode, ultrastretchability

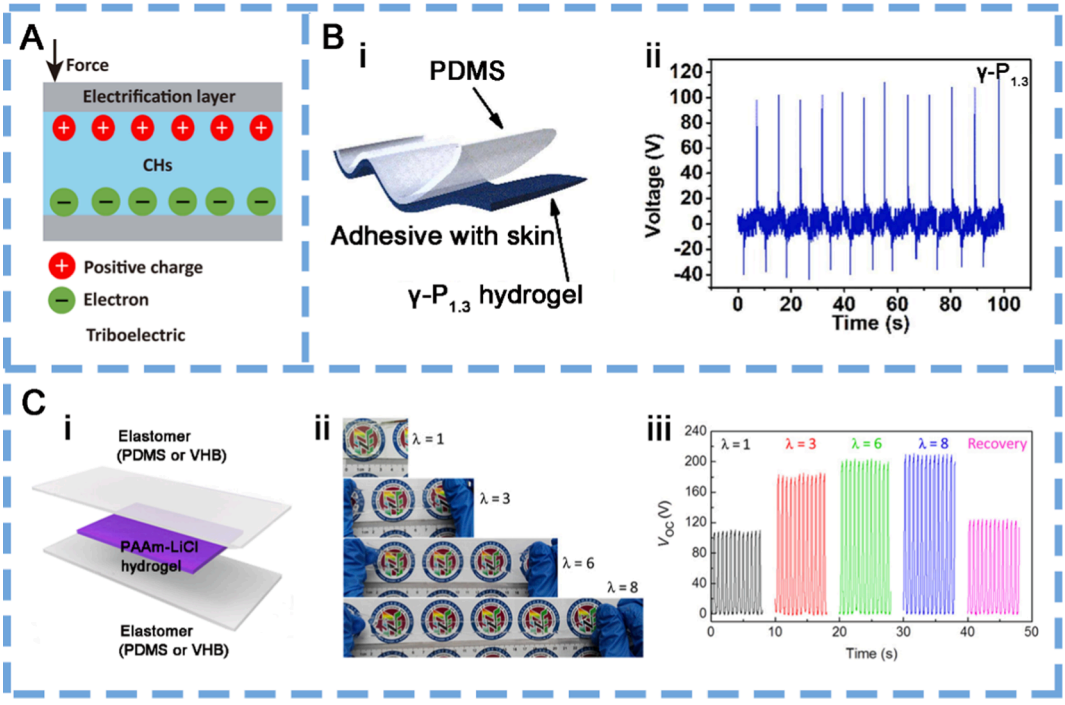

Fig. 11. CHs-based TENGs. (**A**) Working mechanism. (**B**) i) Schematic diagram of the architecture for TENG. ii) Output voltage of TENG. γ-polyglutamic acid/1.3 wt% PEDOT:PSS (γ-$P_{1.3}$). Reproduced from ref. [67] with permission from Elsevier, copyright 2022. (**C**) i) Structure of CHs-based single-electrode TENG for bioenergy harvesting. ii) Images of the TENG at different stretched states and iii) its corresponding output. Reproduced from ref. [301] with permission from American Association for the Advancement of Science, copyright 2017.

over 1160% was realized, and the device showed no degradation under about 8 times stretching (Fig. 11Cii-iii). Other unique properties of CHs, including self-healing capability, good biocompatibility, transparency and adhesivity, have also been utilized in CHs-based TENGs for self-powered sensing and mechanical energy harvesting [304,305].

Since CHs used for TENGs in 2017 [301], numerous papers of CHs-based TENGs have been reported with high output voltage (>250 V) [306], high stretchability (>1000%) [307] and ever-improving power density (maximum 4.22 W/m^2) [298], lighting up hundreds of LEDs in laboratory. However, unlike LEDs that can be lit in each tapping process, portable electronic devices all need continuous power to operate, such as bluetooth, electronic watches, and some sensing devices [43,308]. Therefore, a power management system is required, including rectifiers [305], super-capacitors [309] or batteries [310], to transform the AC current to DC current and to amplify the electrical outputs. Due to their unique properties (stretchability, conductivity, and ionic transport properties), CHs have been ideal frameworks to design and construct flexible supercapacitors and batteries. These self-charging all CHs-based TENGs systems have provided an effective method for long-term, timely, and continuous portable health tracking managements [311,312].

4.2. Piezoelectric nanogenerators (PENGs)

PENGs have been long studied for mechanical energy harvesting. They can convert environmental energies to electrical signals, including human taps, walking steps, and sound waves [313–316]. The displacement of cations and anions under mechanical deformations can lead to the unbalanced charge center of anoins and cations, forming the polarized dipoles. The continuous reversible deformations then further result in the time-variation of the electric fields of the polarized charges, which can therefore produce conduction current in the external circuits and realize the mechanoelectric energy conversion (Fig. 12A).

Common commercial piezoelectric materials are piezoceramics (ZnO, ZnS, InN, GaN *etc.*) that are mechanical strong, chemically inert and rigid, exhibiting excellent piezoelectric properties. Whereas their inherent rigid characteristics make them unavailable for flexible devices [28]. Other types of piezoelectric materials are piezopolymers, including polyamide 11 (PA-11) [317], polypropylene (PP) [318], poly(vinylidene fluoride) (PVDF) [319] and its copolymer poly(vinylidene fluoride-co-tri-fluoroethylene) (PVDF–TrFE) [320], which are inherently flexible. Composites combining these piezoelectric materials in forms of nanofiber, nanosheet and polymer films have been widely used. For example, PDMS with good biocompatibility and flexibility has been used as substrates for wearable PENGs. However, their poor adhesion, unsatisfied softness, and huge difference in modulus with skin still limits their employment in biomechanical and biomedical applications. Moreover, their densely network also result in poor permeability.

Hydrogels with adhesion, unique mechanical properties, and good biocompatibility have been considered as available substrates for PENGs [321–323]. Incorporation with different easy-processing piezoelectric polymers (mainly PVDF) which have good stability

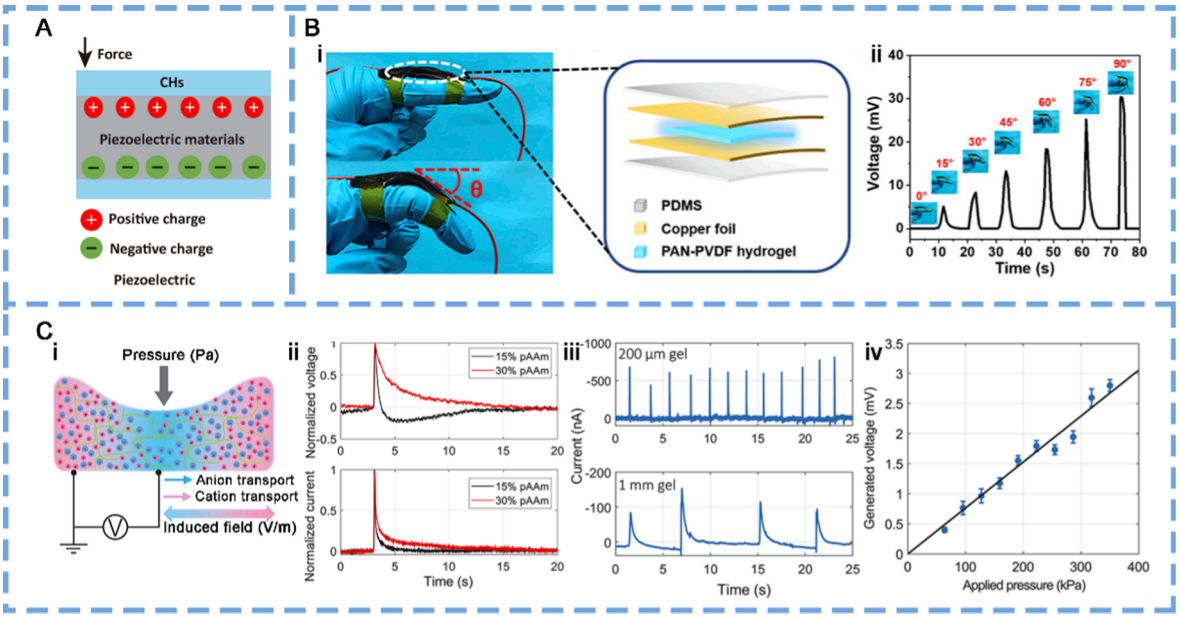

Fig. 12. CHs-based piezoelectric nanogenerators (PENGs). (**A**) Working mechanism of CHs-based PENGs. (**B**) Structure i) of CHs-based PENG for bioenergy harvesting and ii) its output. Reproduced from ref. [327] with permission from American Chemical Society, copyright 2019. (**C**) i) Schematic of a polymer gel under indentation, exhibiting differential ionic displacement and field. ii) Piezoionic transient response under a step compression form 15 and 30% PAAM swollen in 1.5 M NaCl. iii) Piezoionic transient response under repeated step compressions onto a 200-mmthick (top) and a 1-mm-thick (bottom) pAAm gel swollen with 1.5 M NaCl, exhibiting decay times of ~ 50 ms and ~ 5 s, respectively.iv) Peak generated voltage as a function of applied pressure (data are mean ± SD, N = 3). Reproduced from ref. [328] with permission from American Association for the Advancement of Science, copyright 2022.

[324], CHs-based PENGs have been used as mechanical harvesters for wound healing and self-powered sensing [325,326]. PVDF has five different crystal phases, among which the β-phase in PVDF shows largest polarization and piezoelectric sensitivity [28]. For PVDF-dopped CHs-based PENGs, dipole moment in CHs will be rearranged when a certain force is applied, leading to voltage differences on its surfaces. Its output depends on the concentration of the dipole moment and the shape variables produced by the CHs under the force (Fig. 12B) [327]. For CHs-based PENGs, their output shows excellent stability, even after large deformations of stretching and twisting. Meanwhile, PVDF with polymer chains can form a dipole–dipole interaction network to enhance mechanical properties and facilitates the nucleation of the highly electroactive β-phase which can enhance the output of PENGs.

Apart from piezoelectric materials-based CHs-based PENGs, piezoionic hydrogels have also been recently reported (Fig. 12C). The dissolved mobile ions in ionic conductive hydrogels have similar physicochemical properties compared with the ionic solutions. External pressure applied to ionic CHs can displace the mobile anions and cations inside hydrogels. Due to the differences in the diffusivity of cations and anions, there will generate a net ionic current in the hydrogel and therefore the hydrogel is polarized due to the formed concentration gradient of net ion charges along the direction of pressure gradient (Fig. 12Ci). As a result, potential difference will be built up between the two electrodes and current flows through the circuit if it is short-circuited. The pressure-sensing voltage is positive when the anion diffuses faster than the cation; the voltage is negative when the cation diffuses faster than the anion [328]. Furthermore, the devices can be modified easily by changing the thickness of the hydrogels, types (size) or concentration of ions, and the output signal pulse width can be adjusted (Fig. 12Cii-iii). A unique property of piezoionics is that it can output high currents at small voltages, in contrast to electronic PENGs and TENGs (Fig. 12Civ).

The voltage and current output of electronic PENGs are similar with TENGs, whereas the piezoionic generator output high current but low voltage. At present, different energy management systems using capacitors and supercapacitors have been used to enhance their practicality. Meanwhile, using hybrid energy harvesters, such as combed TENGs and PENGs, can also increase their electrical outputs.

4.3. Hydrovoltaics

Water is the most abundant resource with various forms that are essential to our daily life and dominates energy transfer in a variety of natural phenomena, despite the long history of harvesting mechanical energy from water flow [24,329,330]. Generating electricity from direct interaction between nanomaterials with flowing, waving, dropping and evaporating water leads to a new energy conversion effect termed hydrovoltaic effect [331]. Depending on the working mechanisms, wearable hydrovoltaics can be mainly divided into moisture-induced generators (MIGs) and evaporation-induced generators (EIGs). In comparison to TENGs and PENGs, hydrovoltaics have the ability to harness energy from water present in the environment or human body, providing a source of spontaneous and direct current output for self-powered systems [332]. Recent research has highlighted the potential of combining hydrovoltaic nanomaterials with flexible substrates [333–335]. However, a crucial challenge for developing exciting hydrovoltaic devices is ensuring a sustainable water supply, which is not readily available for portable devices. Meanwhile, most of the hydrovoltaic

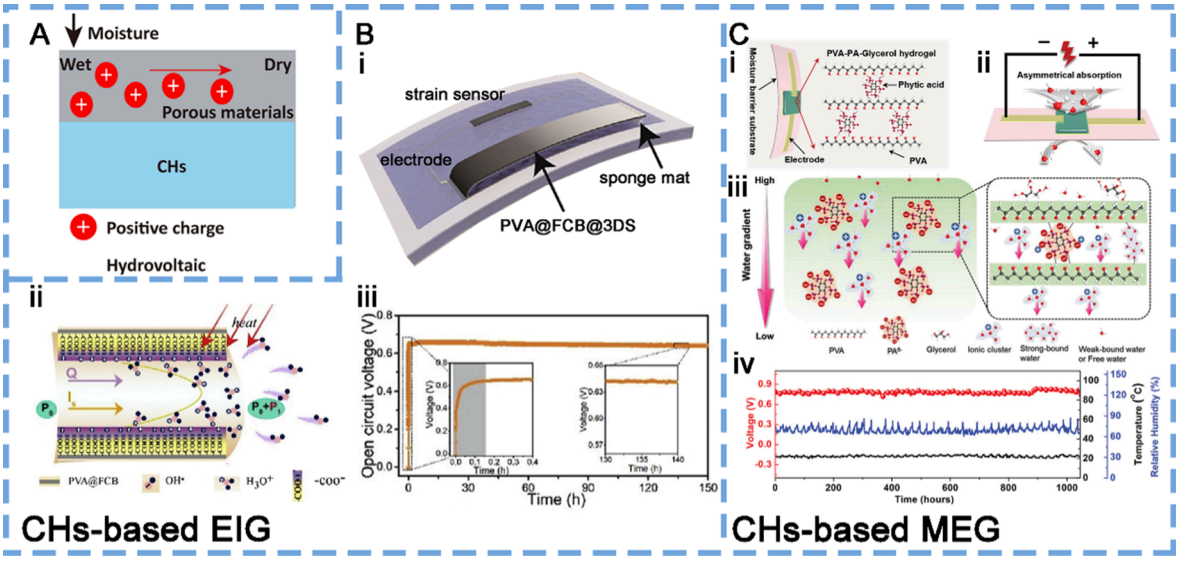

Fig. 13. CHs-based hydrovoltaics (evaporation-induced generators, EIGs; moisture-induced generators, MEGs). (**A**) The working mechanism of CHs-based EIGs. (**B**) i) Schematic diagram, ii) working principle, and electrical output iii) of a superabsorbent hydrogels-based EIG. Reproduced from ref. [342] with permission from Elsevier, copyright 2020. (**C**) i) Structure of a CH-MEG device with asymmetric-moisture penetration layers. ii) Schematic diagram of the asymmetric moisture stimulus-induced potential in the MEG. iii) The proposed mechanism of moisture-induced ionic diffusion process. iv) The continuous voltage output of an MEG device under open ambient environment. Reproduced from ref. [347] with permission from Wiley-VCH, copyright 2022.

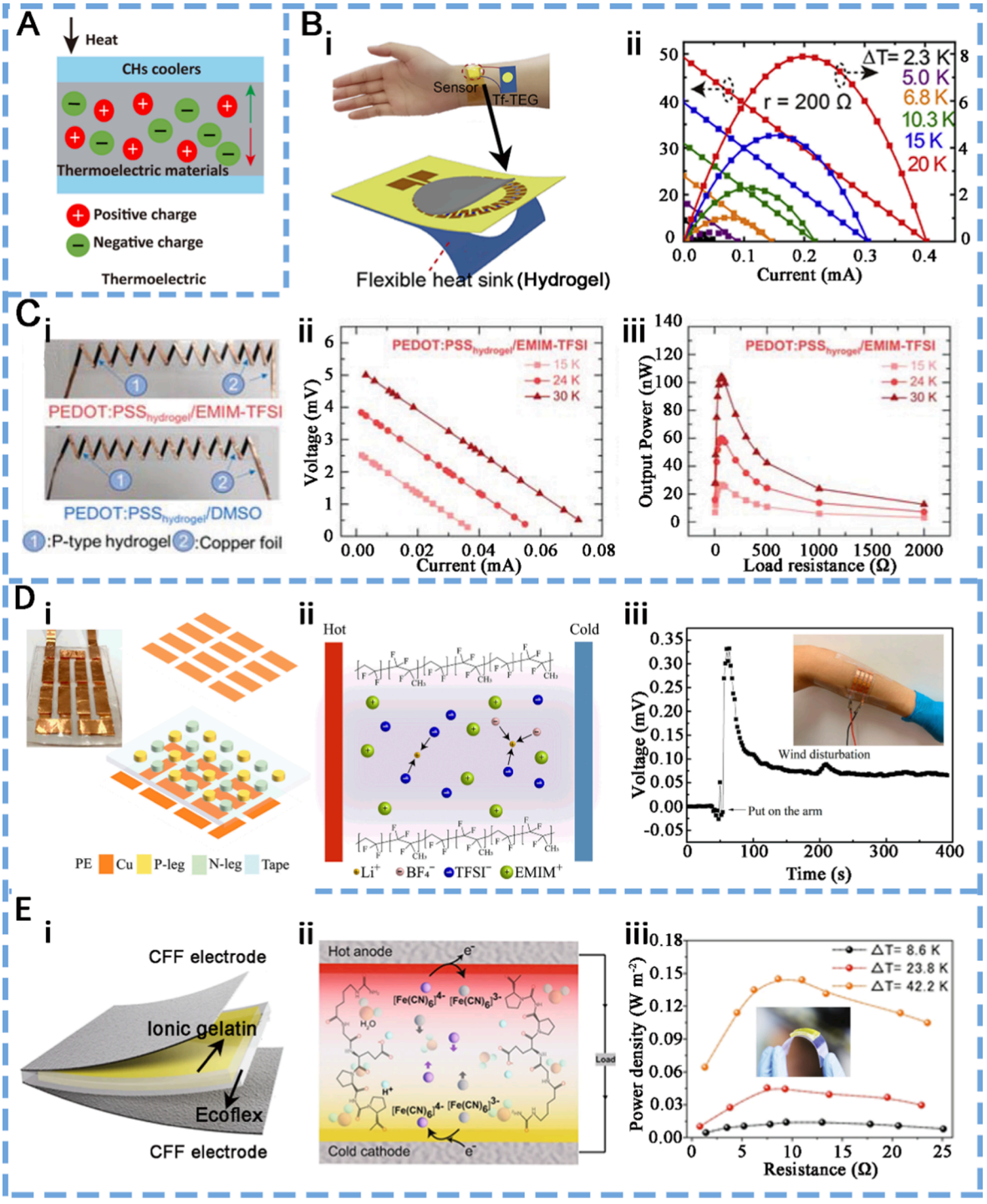

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Fig. 14. CHs-based thermoelectric generators (TEGs). (**A**) Working mechanism. (**B**) i) Schematic illustration a thin-film TEG (tf-TEG). ii) Output of the tf-TEG under different temperatures. Reproduced from ref. [360] with permission from Elsevier, copyright 2020. (**C**) CHs as p-type TEGs. i) Optical images of TEGs made of PEDOT:$PSS_{hydrogel}$/EMIM-TFSI and PEDOT:$PSS_{hydrogel}$/DMSO hydrogels. ii) The voltage-current curves of the TEG at three temperature gradients. iii) The generated power of the TEG as a function of load resistance. Reproduced from ref. [41] with permission from Wiley-VCH, copyright 2022. (**D**) CHs-based i-TEGs. i) A schematic of the design with 12 pairs of p- and n-type ionogels together with the picture of the device. ii) Schematic to illustrate the interaction between Li^+ cations and anions during the thermodiffusion of ions in the polymer channel. iii) Voltage generated from the device on a human arm at 25 °C. Reproduced from ref. [365] with permission from American Association for the Advancement of Science, copyright 2022. (**E**) i) Schematic diagram of the construction of a fabricated TEG. ii) Schematic figure of the diffusion, redox reaction, and interaction of the ions in the fabricated flexible ionic gelatin under a temperature gradient and iii) its power density at different loads. Reproduced from ref. [344] with permission from Springer Nature Limited, copyright 2022.

materials are unstretchable or inflexible, such as semiconductor nanomaterials [336], graphene [337], silicon nanowires [338], and natural wood [339], restricting further wearable applications.

Hydrogels contain large amount of water (maximum over 90%), which can be the distinctive portable water supply for hydrovoltaic devices. Despite ongoing development, hydrovoltaic technology is still in its infancy, particularly in the area of CHs-based devices [340,341]. Typically, specific designed hydrogels with good water management abilities can be used to efficiently collect moisture from ambient atmosphere, transport water vapor for continuous water supply [342], and provide the migration route for protons [40] (Fig. 13A). For EIGs, electricity from water phase change from liquid to gaseous driven by atmospheric heat, as capillary flow caused by evaporation of water at the liquid–solid interface drives charge transport (Fig. 13Bii) [342,343]. To ensure the constant water supply for hydrovoltaic devices, superabsorbent hydrogels have been used to capture water vapor from the environment. These hydrogels were combined with PVA/FCB/DS to generate electricity (Fig. 13Bi). The water inner hydrogels are driven by capillarity through generator film when the nanochannel contains numerous oxygen groups full of water, creating an overlapped EDL between water and nanochannel. Negatively charged channels allow the migration of hydronium ions (H_3O^+), generating a streaming current and higher potential at the top. The potential difference is present over the nanochannels, resulting in output voltage. Note that water was trapped inner hydrogels rather than in a free stage, making this device for long-term portable power supplies (over 150 h, 0.6 V) (Fig. 13Bii). In a more recent report, an ionic hydrogel was used as the heat conduction component between a hydrovoltaic material (dual-size Al_2O_3 layer) and the surrounding environment. This increased evaporation rate and improved output voltage, resulting in a stable output voltage of 6.4 V under sun irradiation, which represents the highest output voltage in a single device achieved in this field so far [344].

For MIGs, electricity comes from the oxygen gradient conformation or water gradient in active materials during water phase change from gaseous to liquid released massive chemical potential energy [345,346]. Currently, hydrogel-based hydrovoltaic devices can achieve electricity generation depending on ionic hydrogel due to its high capability of moisture sorption and fast ion diffusion ability [40]. An efficient ionic hydrogel MEG that can generate a constant voltage of 0.8 V for over 1000 h has been reported (Fig. 13Ci) [347]. With great hydrophilic nature, the ionic hydrogel can capture water molecules from moist air (Fig. 13Cii). Meanwhile, numerous micro- and nanoscale pores of this ionic hydrogel provide a large number of migration paths for ions. Depending on the analysis of 2D-FTIR, when phytic acid (PA) with low dissociation constantly encounters cluster water, its six protons are readily dissociated, forming protonated water clusters (H_3O^+ or H^+ $(H_2O)_n$). Water clusters ions transport as a vehicle to produce a large diffusion current and internal potential (Fig. 13Ciii). Such a CH-based hydrovoltaic device can achieve an output power density of 35 $\mu W/cm^2$, which can be used to power flexible sensors and easy to integrate on a large scale, offering an adequate power source for self-powered wearable devices (Fig. 13Civ).

For both CHs-based EIGs and MIGs, their stretchability and output changes during stretching process has been rarely investigated. Compared with other materials based hydrovoltaic devices, CHs-based hydrovoltaic can be largely deformed and maintain the initial performance. Its unique property is collecting moisture from environment or/and human body fluids and can be easily accessed through capillary or osmotic action for sustained hydrovoltaic process. With the ongoing development of hydrovoltaic technology in recent five years, various active materials have been explored in this field. Compared with TENGs and PENGs, its continuous direct current provides a clear path for portable devices. Nevertheless, it is still far from real-world applications with a low power density. Nowadays, moist environment demands are also crucial factors for both MIGs and EIGs. For constant wearable power supply, CHs-based hydrovoltaic devices could be rationally designed to collect moisture or sweat from human for energy harvesting [348].

4.4. Thermoelectric generators (TEGs)

TEGs have long been an excellent candidate for powering wearable electronics due to their ability to convert heat energy to electric energy through Seebeck effect (the so-called Peltier effect) or Soret effect [32,349]. Thermoelectric device creates a voltage when there is a temperature gradient between two connected conductors or semiconductors. For electronic TEGs (e-TEGs), electrons (holes) as charge carriers driven by electromotive force under temperature gradient, generating a thermopotential along the opposite direction to the gradient (Fig. 14A) [350–352]. Conventional TEGs are based on metal materials that are rigid and brittle due to their good charge mobilities [353,354]. To construct flexible TEGs, thermoelectric films [355], fibers [356], and organic TE materials [357] have been developed.

Hygroscopic hydrogel coolers as self-sustained alternatives for effective heat removal have been researched for several years. Hydrogels can be used as a flexible heat sinks to produce a large temperature gradient for high power generation [358,359]. A type of atmospheric hygroscopic ionic hydrogel has been developed to achieve higher heat dissipation for TEG, allowing the maintenance of a

stable temperature gradient between the environment and human body (Fig. 14Bi). With a temperature gradient of 20 K, the CHs-based e-TEGs produced an output voltage of 78 mV and P_{max} of 7.9 μW (Fig. 14Bii) [360]. As a flexible and stretchable electrode, CHs have been well used for charge transport. An ionic liquid doped PEDOT:PSS CHs with super high conductivity (305 S/cm) has been used in flexible, self-healing, wearable TEGs with low internal resistance [41]. These CHs as p-type stripes were connected by copper foil (Fig. 14Ci), achieving an output voltage of 4.9 mV (Fig. 14 Cii) and a P_{max} of 84.8 nW at a temperature gradient of 30 K (Fig. 14 Ciii).

For ionic thermoelectric generators (i-TEGs), the generation of electric potential is rooted in thermodiffusion and thermogalvanic effect [352]. The thermodiffusion effect in i-TEGs arises from the Soret effect. When a temperature gradient is applied, ions migrate

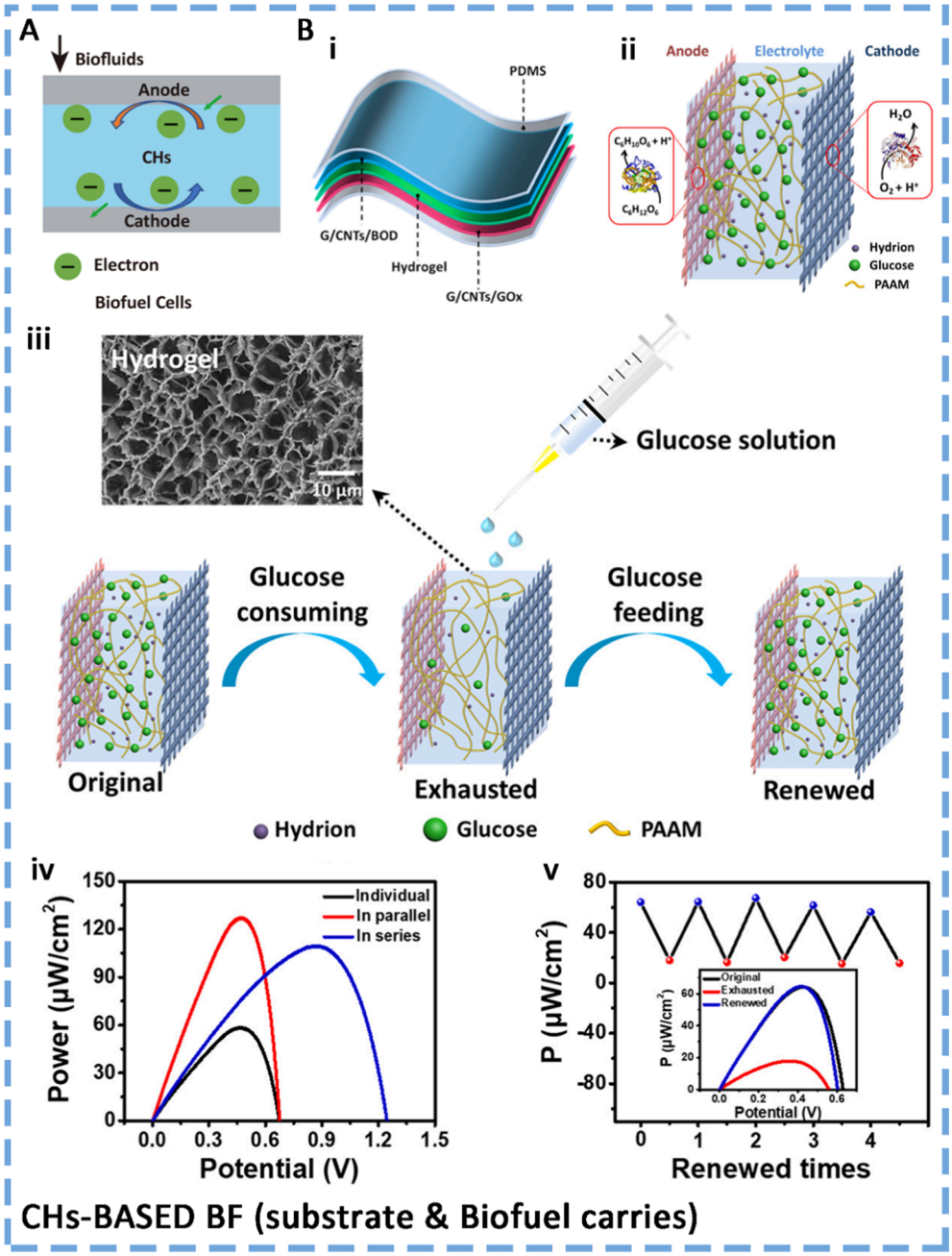

Fig. 15. CHs-based biofuel cells (BFs). (**A**) Working mechanism. (**B**) Schematic of the structure i) and working mechanism ii) of flexible and stretchable BFs. iii) Schematic to show the functions of CHs. iv) Power profiles of two biofuel cells connected in parallel (red curve) and in series (blue curve). v) The renewable performance of BF for four times. Reproduced from ref. [375] with permission from American Chemical Society, copyright 2022.

from the hot side to the cold side, resulting in a difference in ion concentration at the two electrode sides. The CHs should have high ionic transmission rate, high thermopower and excellent flexibility [361–364]. A solid ionic liquid-based ionogels with ultrawide tunable thermovoltage from −15 to 17 mV has been reported [365]. For wearable CHs-based i-TEG, a device with 12 p-n pairs has been demonstrated with a total thermopower of 0.358 V/K, showing potential for human heat generation (Fig. 14 D).

The thermogalvanic is based on thermally driven redox reactions (oxidized at anode and reduced at cathode) [366]. For example, an ionic CHs based thermogalvanic i-TEGs has been proposed, with the redox reaction of reaction $[Fe(CN)_6]^{4-} \rightarrow e^- + [Fe(CN)_6]^{3-}$, achieving a P_{max} of 0.146 W/m^2 at the temperature of 42.2 K (Fig. 14E) [344]. For CHs-based TEGs, their output did not change much after numerous bending cycles. Meanwhile, the unique properties of CHs, such as strong adhesion, fast self-healing, and good biocompatibility give more opportunities for on-skin application. However, compared with metal-based TEGs, the output and stability of CHs-based TEGs still have a long way to go in both charge transport ability and heat skin stability during moisture sorption and evaporation.

4.5. Biofuel cells (BFs)

Biofuel production (non-fossil fuels) has been used long before the oil era in the later 19th century. Compared with other types of cells and energy generators, BFs convert biochemical energy to electricity through redox reaction [367–369]. At the anode, electrons and protons are released from the chemical fuel by the action of the catalyst; while in the cathode region, electrons reduce the electron acceptor (O_2, H_2O_2) and combine with the transferred protons (H^+) from water. Current is created by electrons transferring to the cathode through an external circuit, while protons are transferred to the cathode directly through electrolyte (Fig. 15A). Based on different catalysts, there are three types of BFs: microbial (e.g. microalgae, actinomycetes), inorganic (*e.g.* Pt, Au), and enzymatic (*e.g.* glucose oxidase, D-glucose dehydrogenase) [370,371]. With good biocompatibility, BFs can serve as wearable and implantable energy generators that convert the chemical energy of lactic acid or glucose from human blood, sweat, saliva and other biofluids into electricity [372,373]. For CHs-based BFs, the functions of hydrogels can be various, such as adhesive substrate [374], electrolyte [375] and carriers of microorganisms and biofluids [376].

CHs, with their suitable adhesion and stretchability, can serve as substrates for wearable biosensors and can be attached to human skin directly (Fig. 15Bi). Conventional BFs use aqueous solution as electrolyte, which is a big challenge for avoiding leakage during stretching process [375,377–379]. Hydrogels with excellent liquid holding capacity have been considered as promising candidate

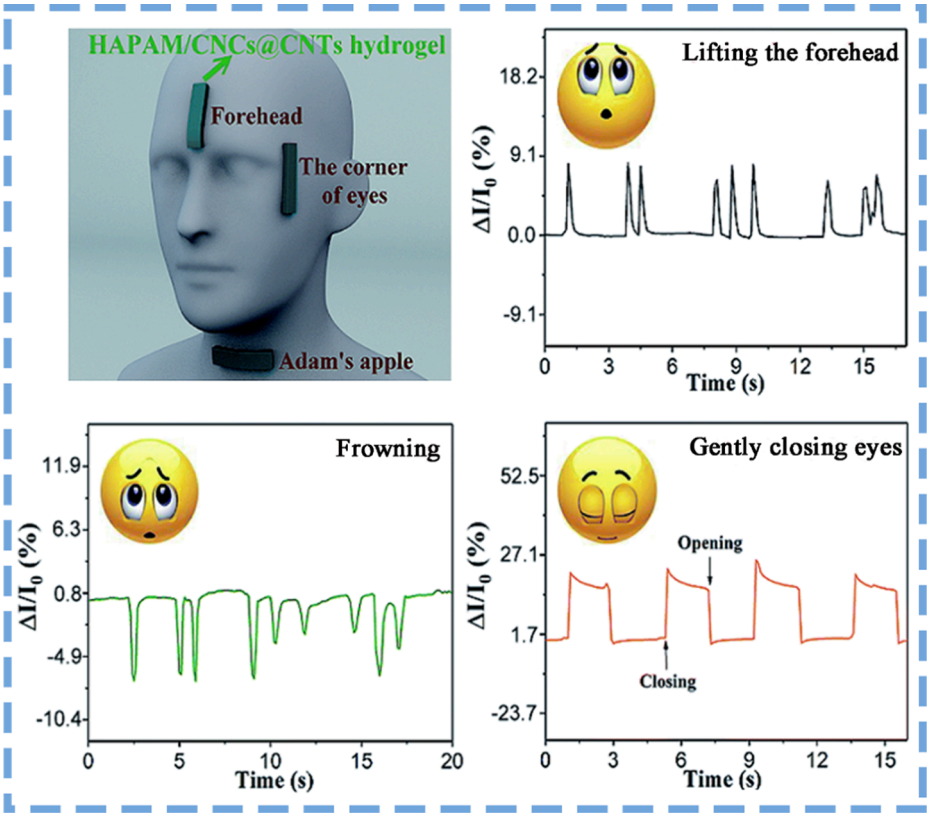

Fig. 16. CHs-based highly sensitive strain sensor attached to the forehead, the corner of the eye to detect facial expressions and corresponding current responses. Reproduced and reprinted with permission. Reproduced from ref. [395] with permission from The Royal Society of Chemistry, copyright 2019.

electrolytes for BFs (Fig. 15Bii). Incorporating flexible electrodes and hydrogels with suitable catalysts enables BFs to become wearable and reusable for long-termed portable power supply [380]. Meanwhile, after several strain and bending-release cycles, the output for CHs-based BFs were almost unchanged. For example, a type of highly flexible and stretchable CHs-based BFs has been proposed, consisting of conductive textile electrodes and CHs electrolyte. This device exhibited a high open-circuit voltage of 0.65 V and P_{max} of 64.2 $\mu W/cm^2$ at a high strain of 60% (Fig. 15Biv-v) [375]. For microbial BFs, hydrogels with suitable inner structures and environments for fungi and algae have been researched [381]. CHs-based BFs can adhere with human skin directly, which is more suitable for different types of biofluids collection, such as sweat or tears, which contain several metabolites that can be used as fuels, to generate usable electrical energy. In the future, CHs-based BFs should be more autonomous in biofuel capture from human sweat or other biofluids.

5. Self-powered devices

Incorporating with different types of bioenergy harvesting design, CH-based self-powered systems have made significant progress in recent years. In this section, we will introduce the typical application of CHs-based self-powered devices in human motion tracking, health monitoring, human–machine interaction, and nerve stimulation. Although multimodal sensing has recently emerged for a wide range of applications including epidermal electronics, robotics, health-monitoring devices and human–machine interfaces, decoupling the input signals for true realization of multimodal sensors is critically important [387,388]. Some comprehensive reviews on this topic can be found elsewhere [389,390].

5.1. Motion tracking

On-skin strain sensors or skin-attachable nanogenerators can track the motion of different parts of the human body. Owing to the tissue-like softness, high stretchability, and conductivity, CHs have been extensively used for the development of self-powered motion-tracking sensors/nanogenerators [302,391–393]. The key requirement for CHs for self-powered motion-tracking applications is high stretchability and toughness under long-term cyclic mechanical loads.

For example, a wearable self-powered human motion sensor made from highly stretchable quasi-solid state hydrogel, which shows 2800% elongation at break and good strain sensitivity (GF = 4, when the strain is 200%) and detects the movement and sound of the human body is reported [394]. Furthermore, using hydrogel as an electrode, a stretchable (900% stretchability) and healable TENG is developed that can detect human motion and pressure [393]. The human knee joint needs to sustain high stresses (4–9 MPa) for 1 million cycles per year. Inspired by the outstanding anti-fatigue-fracture property of human tissue, a highly durable (>20,000 cycle) hydrogel-based strain sensor has been developed by using partially crystalline fiber-like cellulose nanocrystals/carbon nanotubes into a hydrophobic associated polyacrylamide (HAPAM) hydrogel. The resulting sensor has a high gauge factor (GF = 7.63) and can detect a strain as small as 0.2%. This highly sensitive sensor can monitor subtle facial expressions when attached to forehead and the corner of the eye (Fig. 16) [395]. The strain sensitivity is further improved (GF of 25) with an MXene-based hydrogel (M−hydrogel). This GF can be as high as 80 under a compressive strain. This type of strain sensor can monitor facial expression with even higher precision [396]. Li_2SO_4/polyacrylamide/calcium-alginate (Li_2SO_4/PAAm/CA) has been developed as a self-powered wearable strain sensor with high sensitivity and high thermoelectric voltage, which can real-time monitor different types of human motions such as finger, wrist, elbow and knee movement as well as subtle throat movement [394]. However, the majority of the current reports focus on monitoring 2-dimensional motion, extending the capability in monitoring 3-dimensional motion or making sensors multifunctional would be an interesting research direction.

5.2. Health monitoring

CHs have become increasingly popular in the field of biosignal monitoring due to their ability to improve the accuracy and comfort of wearable devices. To monitor the bio-signals such as electrocardiogram (ECG), electromyogram (EMG), and electrooculogram (EOG) continuously for an extended period of time, electrodes should have high conductivity, high mechanical softness, and maintain intimate contact with skin.

Since hydrogel contains mostly water, achieving anti-freezing property in CHs for operation in extremely cold weather is also important [397,398]. Recently, a 3-D printed super-anti-freezing, highly stretchable, and self-adhesive CH is developed that can capture high-quality ECG, EOG, and EMG at an extremely low temperature of −80 °C [399]. The interface between electrode and tissue is critical in practical applications, particularly for weak EEG signals at the microvolt level, which is also easily influenced by deformation, sweating, and motion. To address these issues, CH used to construct EEG electrodes requires low interfacial impedance, good self-adhesiveness, low modulus, excellent flexibility, and high transparency/biocompatibility. These properties improve EEG signal quality, facilitate long-term observation, and ensure electrode safety [400]. To achieve mechanical softness, skin adhesiveness, and high conductivity concurrently, a mechanically interlocked hydrogel–elastomer hybrid has been developed with successful recording of ECG and EMG for several cycles [401].

Although current CHs can achieve intimate contact and detect weak EEG signals, monitoring EEG in hairy scalp and detachment of electrodes after use still remains a problem [402–404]. A biocompatible on-skin paintable conductive biogel has been developed, which showed temperature-controlled reversible phase transition between the fluidic state and viscoelastic gel state, as well as water-triggered removal property to address the aforementioned challenge [404]. They achieved the conformal contact and dynamic compliance of electrodes with hairy scalp, which contributes to the long-term high-fidelity EEG recording over continuous wearing of

electrodes for several days without the need for additional mechanical fixtures to secure the electrodes (Fig. 17A). To mitigate the power requirements, self-powered wearable sensor systems are facile solutions. For example, all ocean-derived catechol-chitosan-diatom hydrogel (CCDHG)-based stretchable, self-healable, and self-powered TENG as a tremor sensor is reported for monitoring Parkinson's disease (PD) [405]. The tremor sensor is a specially designed M-shaped Kapton film and CCDHG-TENG that operates in contact-separation triboelectric mode (Fig. 17B). The feasibility of the sensor has been tested by attaching to the wrist of a real PD patient, detecting three different conditions: normal, minor, and severe tremors. Extending the capability of CHs beyond biosignal

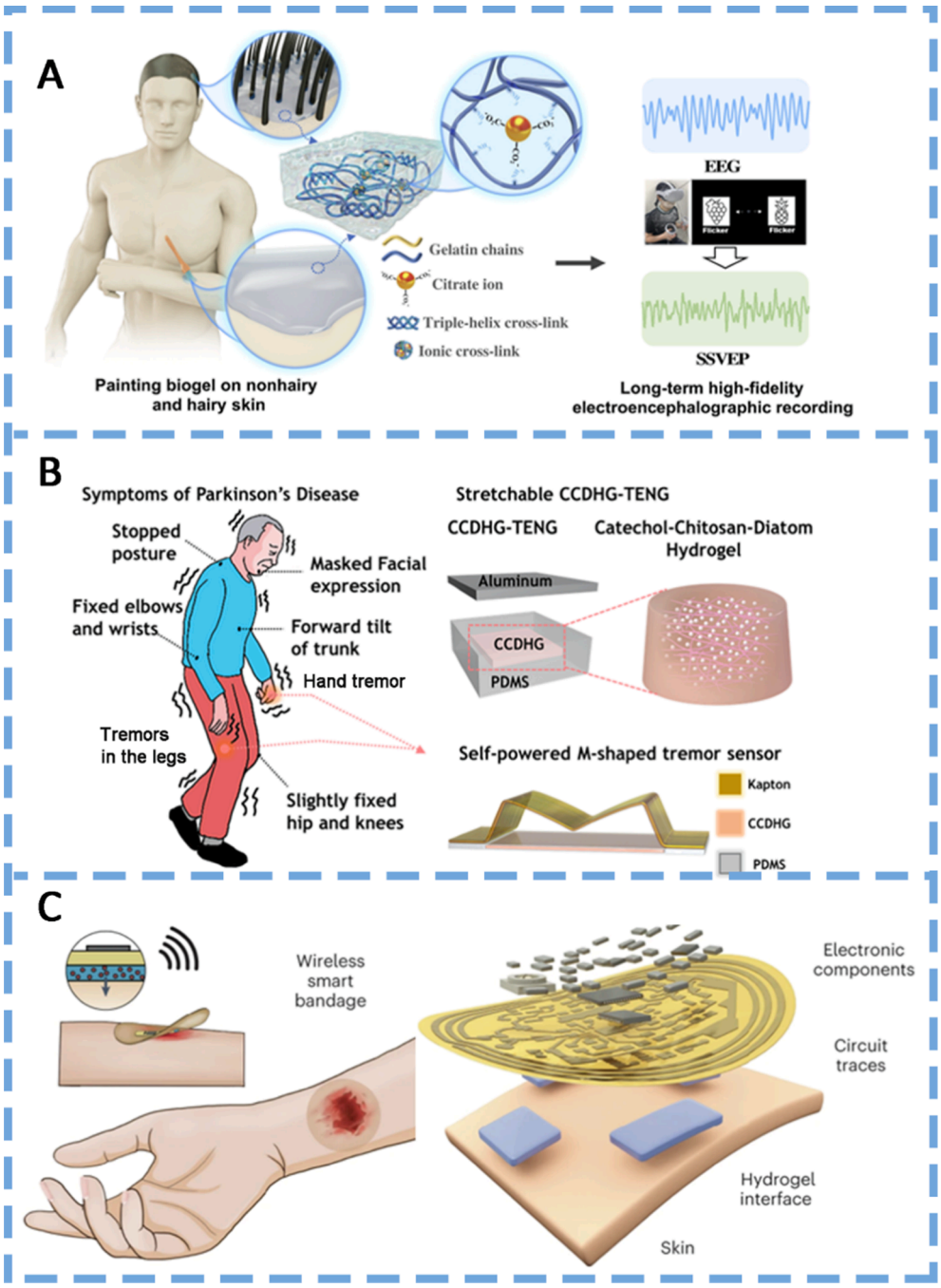

Fig. 17. (**A**) On-skin paintable biogel interface for hairy scalp for high-fidelity EEG recording. Reproduced from ref. [404] with permission from American Association for the Advancement of Science, copyright 2022. (**B**) Catechol-chitosan-diatom hydrogel (CCDHG) conductor based TENG as tremor sensor for Parkinson's disease (PD) monitoring. Reproduced and reprinted with permission. Reproduced from ref. [405] with permission from Elsevier, copyright 2021. (**C**) Wireless smart bandage for wound care and healing comprising of flexible printed circuit board (FPCB) and tissue-interfacing conducting adhesive hydrogel. Reproduced from ref. [409] with permission from Springer Nature Limited, copyright 2022.

monitoring, can enable other healthcare applications such as wound healing [133,195,404,406,407]. Recently, a smart bandage using a hydrogel interface in a closed-loop system that can monitor impedance and stimulate on-demand to accelerate wound healing progress has been developed as shown in Fig. 17C [408]. The battery-free flexible bioelectronic system consists of wirelessly powered sensing and stimulation circuits with tissue-interfacing tough and low-impedance and adhesive hydrogel electrode based on PEDOT:PSS. The resultant group bioelectronic system can heal wound 25% more rapidly than the control.

5.3. Human-machine interaction

The field of human–machine interface (HMI) has become increasingly important in recent years, as technology has advanced to the point where machines can interact with humans in more natural and intuitive ways. The interfaces that enable communication between humans and machines are still primitive, resulting in inefficient and short-term interfacing. These limitations are particularly evident in emerging fields such as brain-machine interfaces, virtual and augmented reality, neuroprosthetics, and medical implants, which require seamless, biocompatible, and long-term communication and interaction. This is because of the inherent mechanical mismatch between current silicon electronics and biological tissues. Although engineering the design of conventional electronics in a thin form factor or utilizing plastic electronics can conform and follow the curvilinear structures of the human body, owing to the tissue-like properties, CHs can bridge the gap and can enable next-generation human–machine interaction seamlessly [146,393,400,410–413].

For instance, a flexible HMI system is developed using organohydrogel TENGs attached to five fingers. Assigning each letter/function of keyboard to one of 2^5-1 variations for 5 fingers tapping individually or simultaneously can mimic the function of keyboard and can write meaningful words. Thanks to the anti-freezing property of the designed hydrogel, this interactive system can operate at −30 °C [393]. A glove-based human–computer interaction (HMI) system using a chemically cross-linked polyacrylamide (PAM)/

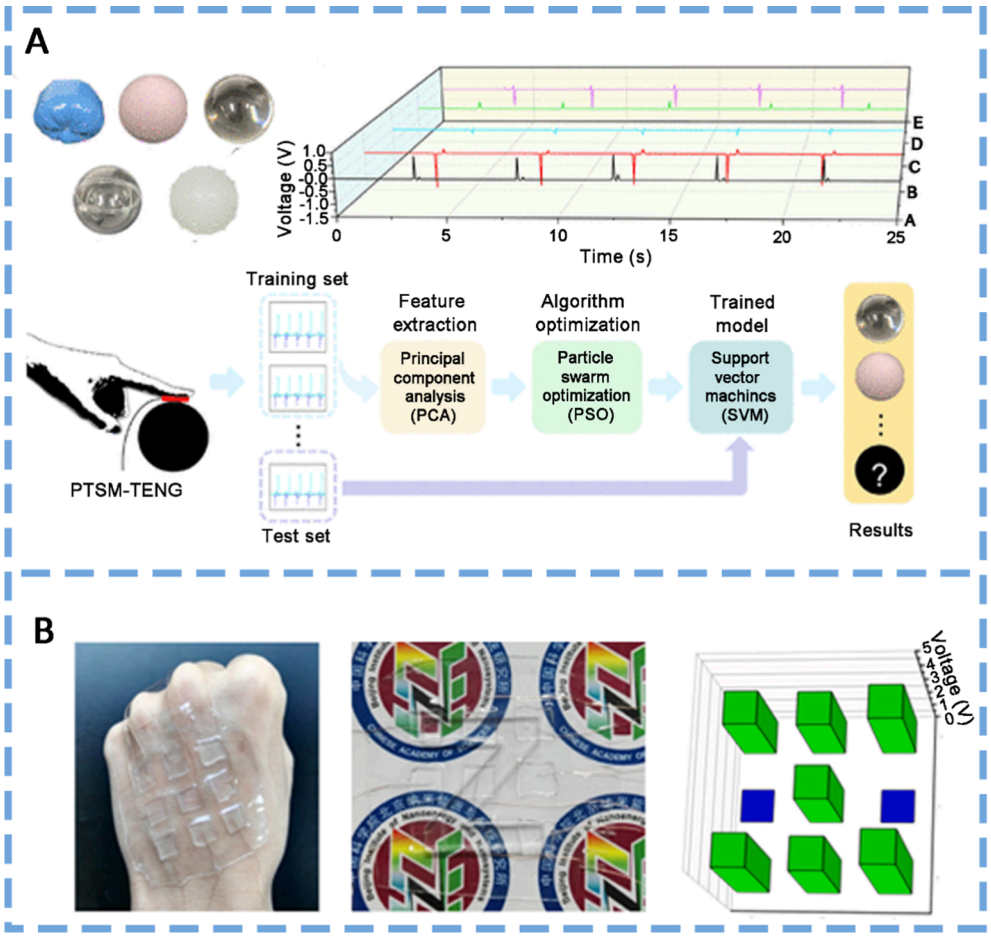

Fig. 18. (**A**) Glove-based human–computer interaction (HMI) system using ionic hydrogel-based TENG. The hydrogel TENG-based glove achieve five spherical object classification and recognition with the help of machine learning techniques. Reproduced from ref. [400] with permission from American Chemical Society, copyright 2023. (**B**) Soft skin-like triboelectric nanogenerator (STENG)-based tactile sensor with 3 pixel × 3 pixel attached on a curvy hand and its tactile operation when pressed with a z-shaped acrylic plate. Reproduced and reprinted with permission. Reproduced from ref. [301] with permission from American Association for the Advancement of Science, copyright 2017.

tannic acid (TA)/sodium alginate (SA)/MXene DN hydrogel (they name it, PTSM hydrogel) based TENGs for hand pose tracking has been developed. Utilizing a machine learning algorithm, the PTSM-TENG can classify and recognize five different spherical objects (Fig. 18A) with an accuracy of 98.7% [400]. Utilizing ionic hydrogel as electrode and electrification layer, a soft skin-like TENG-based tactile sensor system that detects both touch and pressure has been developed. Fig. 18B shows the optical photo of a transparent TENG-based tactile sensor with 3 by 3 pixel attached to a curvy hand. Each pixel generates a proportionate voltage when pressed with a z-shaped acrylic plate [301]. The same group later developed conducting MXene/polyvinyl alcohol (PVA) hydrogel TENG (MH-TENG) based touchpad with great sensitivity, reliability, and identifiability. Utilizing machine learning and subsequent processing, the touchpad can serve as a self-powered written text recognizer and an encryptor for confidential information [304].

5.4. Nerve stimulation

The nervous system is responsible for real-time information transmission between the brain and the body. Any damage to nerve tissue leads to chronic pain, neurological disorders, and disability. Nerve tissues are soft, elastic, and reconfigurable. The stimulation of the nerve can broadly be categorized as non-invasive where electrodes are attached to the skin and perform stimulation, and invasive where electrodes are implanted inside the body usually inside the brain. In either case, ionic currents are injected into the electrolytic medium to excite or inhibit neurons, thereby changing the membrane potential. Since the implantation of electrodes results in scar tissue formation around the electrodes, the interfacial impedance increases significantly which in the long run decreases the stimulation efficacy. It has been reported that reducing mechanical mismatch across electronics-tissue interface elicits significantly less adverse immune responses for chronic implantation [414,415]. CHs having tissue-like properties (Young's modulus < 10 kPa) have gained much attention in bioelectronic nerve stimulation in recent years.

Another key advantage of CHs over conventional metallic electrodes is their superior adhesion properties that can provide reliable contact to nerve tissue and electrodes. A light-stimuli-responsive, highly stretchable conducting polymer hydrogel (CPH) has been synthesized and demonstrated it as an implant for peripheral nerve injury in vivo [416]. A flexible electrical patch (ePatch) using silver nanowire and methacrylated alginate (MAA) to formulate conducting hydrogels has been designed for accelerated wound healing [404]. An interactive wound dressing composed with carbonized polydopamine/polydopamine/polyacrylamide and electroactive

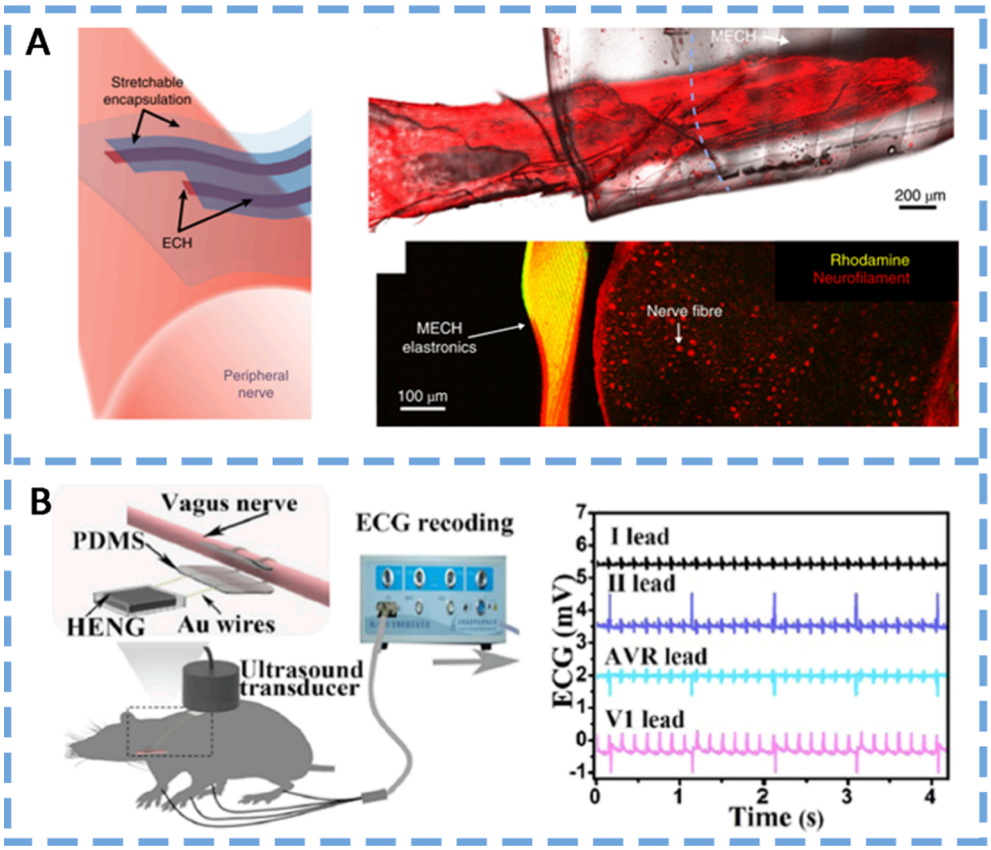

Fig. 19. (**A**) Soft and elastic hydrogel-based microelectronics wrapping sciatic nerve of anesthetized mouse and low voltage neural stimulation in vivo. Reproduced from ref. [168] with permission from Springer Nature Limited, copyright 2019. (**B**) Wireless electrical stimulation of the vagus nerves by ultrasound-responsive programmable hydrogel nanogenerators and 4-channel measurement of rats' electrocardiography (ECG). Reproduced and reprinted with permission. Reproduced from ref. [419] with permission from Elsevier, copyright 2021.

electrospun poly(vinylidene fluoride) (PVDF) membrane to not only deliver electrical stimulation but also to provide an optimal environment for faster wound healing [417].

While most of the epidermal hydrogel-based stimulating devices are used for wound healing [417,418], implantable conducting hydrogels also gained much interest for stimulating neurons [7,419–424]. An electrically conductive hydrogel (ECH) based microelectronics has been developed and engineered (Young's modulus in kPa range) for localized low-voltage electrical stimulation of the sciatic nerve in live mice (Fig. 19A). A general strategy to directly pattern a stretchable insulation layer with microscale resolution using ECH with high aqueous stability and biocompatibility also been proposed. Employing micropatterned ECH as both contact electrode and interconnect reduced the overall impedance 10 times compared to Au use as interconnects and enabled localized low-voltage neuromodulation [168].

The new generation neurostimulators need to be programmable, battery-free, and minimally invasive. To meet these requirements, wireless-powered, battery-free vagus nerve stimulators that utilize implantable high-performance hydrogel nanogenerators (HENGs) and can be activated remotely by ultrasound pulses (Fig. 19B) has been developed. The HENGs are highly efficient and can be used for anti-inflammatory therapy in sepsis by stimulating the vagus nerve [419].

6. Outlook

CHs have undergone significant advancements and are now being widely utilized in the fields of flexible devices and healthcare systems. The cutting-edge research outcomes on CHs have been reviewed, highlighting their distinctive characteristics that make them suitable for use in flexible and stretchable devices. Moreover, this review sheds light on the vast applications of CHs in bioenergy harvesting and self-powered sensing. Despite the remarkable progress in the field of flexible bioelectronics, there is still a considerable gap between research studies and practical applications. While CHs-based self-powered sensing devices can cater to the needs of portable sensing, a number of challenges need to be addressed in terms of materials properties and devices design. To bridge this gap, further research and innovation are needed to explore the full potential of CHs-based devices.

6.1. Balance of mechanical property and conductivity

Mechanical properties and conductivity are two of the most critical aspects of CHs. However, it is a great challenge for CHs to achieve both high conductivity and good stretchability as most electronic conductivity materials cannot effectively dissipate strain energy. Incorporating these conductive materials with another stretchable polymer network forms a stretchable network for CHs, but resulting in a huge decrease in electrical conductivity. Such a compromise between stretchability and conductivity is attributed to the low content of conductive materials after the introduction of another network. To balance these two important properties for CHs, some state-of-the-art methods include acid treatment [169], bi-continuous networks structural designs [425], laser-induced phase separation [11], and ionic liquid doping [41] to compose CHs with high conductivity and suitable stretchability for multiple applications.

6.2. Controllable swelling

Due to the rich hydrophilic groups of CHs. Uncontrollable swelling during water sorption and desorption is still a big challenge for CHs, especially for bioelectronics application in vivo. For example, mechanical deformation of the swelling hydrogels in aqueous conditions may cause a detachment with human tissue, leading to undesirable sensing failure. Solvent replacement [426], by adding hydrophobic-conjugated materials [427] and doping different concentrations [428]of salts into hydrogel matrix, can improve the density and uniformity of cross-linking points without breaking the hydrogel components and non-covalent interactions, which can be used to construct anti-swelling CHs.

6.3. Environmental adaptability

Hydrogels are soft materials that contain large amounts of water. In such composition, water content determines the properties of CHs. For most of existing CHs suffer from turning rigid and poor stretchability in the dry and freezing environment due to their unsatisfying water retention and high freezing point. The multiple demands of water content and the other abilities (self-healing, stretchability, adhesive, *etc.*) for CHs working for a long time in different environments are challenging. Incorporating anionic [429] or ionic liquid [430] to form hydrogels, making CHs moderately water-locked, higher stretchability, conductivity, and lower freezing point to meet the needs in harsh conditions.

6.4. Sustainable power supply

The output of CHs based energy harvesting devices are generally hard to be directly utilized as power sources. The outputs of most TENGs are alternating signals with high voltage (ranging from tens to hundreds of volts, or even higher) and low-current (from nA to μA level). Hydrovoltaics and TEGs generate direct current with voltage of around 1 V and current of a few μA. Common drawbacks of these devices in electrical outputs include relatively low power density (usually at or lower than $\mu W/cm^2$) and fluctuations accompanied with the energy sources. An exciting future direction of CHs-based energy harvesters is incorporation of multiple bioenergy harvesting techniques into one device for further applications. Integrating different kinds of bioenergy harvesting techniques on the

same platform could not only increase the output, but also may compensate for the intermittence of single types. For example, TENGs can only perform self-powered sensing or energy harvesting with human motion. When the wearers are asleep, mechanical energy is minimal, then body heat or/and moisture can be mainly used to generate electricity. [344]. For another part, developing CH-based energy-storage devices such as batteries and supercapacitors incorporated with CHs-energy-generators to achieve self-charging power sources would move these devices a step closer to practical application [312].

6.5. Devices design

The design of CHs-based wearable bioenergy harvesters for self-powered applications typically includes multi-layer that can maintain efficiency under repeated mechanical deformation. In such devices, CHs are much stretchable than other thin layers, which can cause interface mismatch or crack of active layers. For example, dielectric tribo-electrification materials (such as PDMS and PTFE) for CHs-based TENGs and the hydrovolatic materials (Al_2O_3 and carbon black film) for CHs-based hydrovoltaic devices are much thin thinner than CHs layer and will experience much strain at mechanical deformation. The mismatch between different types of layers and CHs must be taken into account. Using a bridge-island structure [431] or reducing the thickness of CHs film [203] can increase the flexibility and the adherence to human body for more sensitive monition. Furthermore, for existing CHs-based self-powered devices, it is reasonable to develop a wireless data transmission. Although several approaches to wireless self-powered applications have been proposed, more flexible and versatile data transmission for compatibility with CHs-based system systems should be pursued in the future.

6.6. Further applications

CHs-based self-powered sensing with unique mechanical flexibility, biocompatibility and good adhesion is a valuable technique that allows direct signal acquisition without extra power supplies or sensors. Though existing literature seem to prove that CHs-based self-powered devices can provide useful data for biomechanical diagnosis, treatment and human–machine interaction, most of them are too early for real-world application. For example, a CHs-based human motion tracking sensor works on the sine-like curves induced resistance change for motion detections in different parts of human body. However, such curves are confused for analyzing the correct part motion. Machine learning can potentially play an important role in extracting information from CHs-based self-powered sensors that is clinically useful and easy to interpret [405].

In summary, the exceptional properties of CHs have demonstrated remarkable potential in the realms of bioenergy harvesting and self-powered applications. Moving towards the future, self-powered CHs-based electronic devices are expected to become more intelligent, wireless, and multifunctional, enabling timely and effective capture and analysis of bio-signals. This comprehensive review provides a profound understanding of the unique characteristics of CHs, fostering a tighter interaction between the realms of natural and artificial. By harnessing the full potential of CHs, researchers can pave the way for the development of novel technologies that have a significant impact on the healthcare industry. The integration of CHs into wearable electronic devices is expected to revolutionize the way we approach healthcare. We hope this review serves as a crucial stepping stone towards unlocking the full potential of CHs and building a more interconnected and efficient self-powered system.

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Declaration of Competing Interest

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

Data availability

No data was used for the research described in the article.

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