Seamless integration of conducting hydrogels in daily life: from preparation to wearable application

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Abstract

Conductive hydrogels (CHs) have received significant attention for use in wearable devices because they retain their softness and flexibility while maintaining high conductivity. CHs are well suited for applications in skin-contact electronics and biomedical devices owing to their high biocompatibility and conformality. Although highly conductive hydrogels for smart wearable devices have been extensively researched, a detailed summary of the outstanding results of CHs is required for a comprehensive understanding. In this review, we summarize the recent progress in the preparation and fabrication of CHs for smart wearable devices. Improvements in the mechanical, electrical, and functional properties of high-performance wearable devices based on CHs that can be seamlessly integrated into our daily lives are reviewed.

Keywords: Conducting hydrogels, wearable electronics, mechanical properties, biomedical applications



Figure 1. Overview of CH development, including their preparation, fabrication, functionality improvement, and applications. Reproduced with permission.^[17] Copyright 2022, American Chemical Society; 2020, Wiley-VCH; 2020, Springer Link; 2016, Wiley-VCH; 2022, Elsevier.



Figure 2. In-situ preparation of CHs. (a) Mechanism of in-situ polymerization of PPy on a PAm/CS hydrogel. Reproduced with permission.^[26b] Copyright 2018, American Chemical Society. (b) In-situ preparation of anisotropic PVA/PANI hydrogels. Reproduced with permission.^[7a] Copyright 2020, Springer Nature. (c) Formation of pure PANI hydrogel from aniline and ATMP monomers using post-polymerization. Reproduced with permission.^[27a] Copyright 2016, Springer Link.



Figure 3. Preparation of non-CP-based CHs. (a) Illustration of preparation of CHACC-LM hydrogel through chemical crosslinking. Reproduced with permission.^[18] Copyright 2022, Wiley-VCH. (b) Schematic diagram of preparation of Tet-TA/GO hydrogels through horseradish-peroxidase-catalyzed crosslinking. Reproduced with permission.^[52] Copyright 2015, Royal Society of Chemistry. (c) Preparation of Zn-dendrite-free CHs. Reproduced with permission.^[53] Copyright 2022, Wiley-VCH.



Figure 4. (a) Schematic illustration of DIW 3D printing of PEDOT:PSS through lyophilization and redispersion process. Reproduced with permission.^[76a] Copyright 2020, Springer Nature. (b) DIW 3D printing of Pluronic F127-dimethacrylate via micelle formation and additional ionic bond formation between polyacrylic acid (PAAc) and iron ions. Reproduced with permission.^[11a] Copyright 2021, Wiley-VCH.



Figure 5. Schematic illustration of the SLA 3D printing of (a) Am-based CHs in potassium chloride (KCl) along with the chemical structure of the precursor solution. Reproduced with permission.^[77a] Copyright 2022, Elsevier. (b) 3D PANI hydrogels produced from aqueous precursor solution and their chemical structure. Reproduced with permission.^[77b] Copyright 2021, Royal Society of Chemistry. (c) Micropattern of photocurable PEDOT:PSS with polyethylene glycol diacrylate (PEGDA). Reproduced with permission.^[77c] Copyright 2019, Elsevier.



Figure 6. Schematic illustration of DLP printing. (a) Microemulsion processing of P(UA-*co*-Am-*co*-AAc) hydrogel and SEM images for resulting (left) cured hydrogel and (right) hydrogel post-processed with ferric ions. Reproduced with permission.^[78a] Copyright 2021, American Chemical Society. (b) Formation mechanism of PAm-PEGDA hydrogels. Reproduced with permission.^[78b] Copyright 2019, Royal Society of Chemistry. (c) Scheme showing the mechanism of microreactive inkjet printing with PEDOT:PSS and EMIM:ES. Reproduced with permission.^[79] Copyright 2019, American Chemical Society.



Figure 7. Schematic illustration of (a) microfluidic preparation of hydrogel microfiber containing CNTs and resulting double-network structure. Reproduced with permission.^[80b] Copyright 2020, American Chemical Society. (b) (left) Solvent exchange of poly(*N*-isopropylacrylamide-*co-N*,*N*'-diethylacrylamide/chitosan) (poly(NIPAm-*co*-NDEAm/CS) hydrogel into ionic microfiber, and (right) photographs of LED power source connected with microfiber stretched to various lengths. Reproduced with permission.^[66] Copyright 2022, Wiley-VCH. (c) Illustration of electrospinning of the sandwich structure comprising layers of PVA–aramid nanofibers (ANF) and a layer of silver nanowires (AgNWs)/PVA. Reproduced with permission.^[44] Copyright 2021, Wiley-VCH.



Figure 8. Approaches used to improve hydrogel functionalities. Reproduced with permission.^[101] Copyright 2020, Elsevier; 2023, Elsevier. 2020, Elsevier, and 2022, Springer Link.

Improved properties	Strategy	Type of hydrogels	Constituents	Improved parameters	References	
Mechanical	NPs as crosslinking agents	Polymer NP- PANI-g-MeGC crosslinked hydrogel Nps PAm		Elongation at break = 400%	[102]	
		NP-crosslinked hydrogel	DACS Nps Collagen	Tensile modulus = 6.5 kPa Structural stability = compression up to 200 g	[102]	
		Non-covalent micelle crosslinked hydrogel	PEG, UPy	Elongation at break = 1000% Tensile strength = 240 kPa Tensile modulus = 23 kPa of Compressive strength = 4MPa	[103]	
	Interpenetrating polymer networks	Physical crosslinked hydrogel	PEDOT:PSS, PANI, Phytic acid	Compressive strength = 41.6 KPa	[104]	
		Double-network hydrogel	GelMA, ODEX rGO	Compressive strength = 200 kPa	[100]	
Electrical conductivity	Electronic conductivity: Polymer-based CHs	Physical crosslinked hydrogel	PEDOT:PSS PANI Phytic acid	Energy density= 0.25 mWh cm^{-3} Power density = $107.14 \text{ mW cm}^{-3}$ Areal capacitance = 242.2 mF cm^{-2} Volumetric capacitance = 3.5 Fcm^{-3} Capacitance retention ratio = 80.8%	[104]	
	Electronic conductivity: Carbon-based CHs	Double-network hydrogel	GelMA, ODEX rGO	Ionic conductivity= 2.36×10^{-2} S/m	[100]	
	Electronic conductivity: MXene CHs	Nanocomposite hydrogel	PVA MXene	Performance as electrode = 0.5–2.0 Hz Durability with cycling = 3000 at 50% strain Self-healing efficiency = 15 s as electrode	[64]	

Table 1. Summary of strategies followed to improve various hydrogel properties.

	Ionic conductivity: Electrolyte-based hydrogel	Ionic hydrogel	OSA CMC AGO LiCl	Ionic conductivity = 1.15 S/m	[101]
Other properties					
Adhesion	Redox activity of catechol/quinone groups	Catechol functionalized alginate (C-Alg) CHs	EDC:NHS:Alginate DA Graphene	Adhesion energy: 1.79 J m ⁻²	[12b]
		CHs	PEDOT LS-PAm	Adhesive strength: 20–23 kPa	[105]
Self-healing	Formation of imine bonds (Schiff base)	Polysaccharide- based conductive ionic hydrogel	OSA CMC AGO LiCl	Self-healing efficiency: 90% after 24 h	[101]
Freezing and drying tolerance	Hydrogen bonds between glycerin and water molecules	Conductive ionic PGT Organohydrogel	PAAc:gelatin TA in a water/glycerin AlCl ³⁺	Original mechanical and conductive properties after 7 days at -14°C	[67]
	Hydrogen bonds between glycerol and water molecules	Double-crosslinked CHs	PAAc:CMCs Ca ²⁺ Water and glycerol	Reduction in conductivity (50%) and mechanical properties (20%) after 8 h at -20 °C	[106]
Antibacterial activity	Addition of polymer NPs as crosslinking agents	NP-crosslinked hydrogel	PANI-g-MeGC Nps PAm	Against Staphylococcus aureus	[102]
	Addition of NPs	Non-covalent crosslinked hydrogel	PDA@Ag NPs PANI PVA	Against Staphylococcus aureus and Escherichia coli	[107]
Biocompatibility	Addition of polymer NPs as crosslinking agents	NP-crosslinked hydrogel	PANI-g-MeGC Nps PAm	Viability of over 80% with NIH 3T3 cells	[102]

Multiple non-covalent	Non-covalent	PAm, CECT	Adhesion strength on pig skin = 113.2	[108]
crosslinking	crosslinked hydrogel		kPa	
High-density micelle	Non-covalent	PEG	Adhesion strength on pig skin = 8 kPa	[103]
crosslinking	crosslinked hydrogel	UPy	Mouse fibroblasts cell (L929) viability	
			over 95%	

AGO= Agarose, C2C12= myoblast cell line, CMC= Carboxymethyl chitosan, CECT= Carboxyethyl chitin, DACS= Dialdehyde cholesterol-modified starch, GelMA= Gelatin methacrylate, GF= Gauge Factor, LS= Sulfonated lignin, L929= Mouse fibroblasts cells, MeGC= Methacrylated glycol chitosan, MXene= Transition-metal carbide/nitride, ODEX= oxidized dextran, OSA= Oxidized sodium alginate, PANI= Polyaniline, PAm= Polyacrylamide, PVA= Poly(vinyl alcohol), PANa= Sodium phytate, PDA@Ag NPs= polydopamine decorated silver nanoparticles, PEDOT:PSS= Poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate), PEG= poly(ethylene glycol), PPY= Polypyrrole, rGO= reduced graphene oxide, SA= Sodium alginate, UCMSCs= Umbilical cord mesenchymal stem cells, C2C12= myoblast cell line, UPy= Ureido Pyrimidinone, Nps= nanoparticles



Figure 9. (a) Photographs of PAm and PANI NPs crosslinkers in PANI NPs-PAm hydrogels along with an SEM image. Reproduced with permission.^[102] Copyright 2022, Wiley-VCH. (b) Illustration of P(Am-Hb/CM) and the dissipated energy at various strains. Reproduced with permission.^[116] Copyright 2022, Elsevier. (c) Schematic illustration of tensile fracture in PVA samples, including the preparation procedure and tensile-stress curves. Reproduced with permission.^[117] Copyright 2022, Elsevier.



Figure 10. (a) Schematic of strain sensor based on brick mortar structure and CHs. Reproduced with permission.^[8b] Copyright 2019, Royal Society of Chemistry. (b) Piezoresistivity mechanism of a PVA-PPy strain sensor in response to small strain. Reproduced with permission.^[135] Copyright 2020, American Chemical Society. (c) Strain sensors based on PVA/MXenes material and examples of signature sensing, handwriting, voice sensing, and changes in resistance in response to similar words. Reproduced with permission.^[136] Copyright 2018, American Association for the Advancement of Science.

Hydrogel	Network structure	Open-circuit voltage (V)	Short-circuit current	Peak power density	Ref.
PVA	SN	200	22.5 μΑ	-	[151]
PAm	SN	312	32.4 µA	2.7 W/m^2	[152]
Cellulose	SN	187	0.51 µA	-	[42]
PAAc	SN	180	65 µA	$625 \ \mu W/m^2$	[153]
НА	SN	20	0.4 µA	5.6 mW/m^2	[8a]
PAm/HEC	MN	285	15.5 μA	626 mW/m^2	[154]
PAm/PDA	MN	230	12 µA	-	[155]
PVA/SA	MN	204	18 µA	0.98 mW/m^2	[149b]
PAm/Alginate	MN	70	0.5 μΑ	135 mW/m^2	[156]
PAm/Cyclodextrin	MN	95	10 µA	0.64 mW/m^2	[157]
PAm/clay	NC	89	16 µA	710 mW/m^2	[3]
PVA/MXene	NC	230	1.2 µA	$0.33 \ W/m^2$	[158]
PAm/Graphene	NC	40	1.6 mA	0.3 W/m^2	[159]
Cellulose/ZnO	NC	58	5.78 µA	42 mW/m^2	[160]

Table 2. Hydrogel electrode materials and output performance of TENGs.



Figure 11. (a) Schematic of a self-powered, smart elastic-band system including the signal acquisition and processing system, and a real-time output interface displaying the potential of this TENG for human-motion monitoring. Reproduced with permission.^[161] Copyright 2021, American Chemical Society. (b) Demonstration of the smart-skin device with the image and digital resolution of light-emitting letters "CityU" and the electronic circuit of the self-charging system used to power an electronic watch. Reproduced with permission.^[153] Copyright 2019, Wiley-VCH. (c) Schematic representation of accelerated wound healing owing to secretion of biomolecules and formation of new tissues under a self-powered device using ionic TENG. Reproduced with permission.^[163] Copyright 2021, Elsevier.

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