

View Article Online
View Journal

Journal of Materials Chemistry C

Materials for optical, magnetic and electronic devices

Accepted Manuscript

This article can be cited before page numbers have been issued, to do this please use: J. Yang, Y. Liu, W. Yan, P. Zhou, Z. Wang, Y. Wang, Y. Zhang, F. Mo, Z. Ji, H. Haick and Y. Wang, *J. Mater. Chem. C*, 2025, DOI: 10.1039/D5TC01896J.



This is an Accepted Manuscript, which has been through the Royal Society of Chemistry peer review process and has been accepted for publication.

Accepted Manuscripts are published online shortly after acceptance, before technical editing, formatting and proof reading. Using this free service, authors can make their results available to the community, in citable form, before we publish the edited article. We will replace this Accepted Manuscript with the edited and formatted Advance Article as soon as it is available.

You can find more information about Accepted Manuscripts in the <u>Information for Authors</u>.

Please note that technical editing may introduce minor changes to the text and/or graphics, which may alter content. The journal's standard <u>Terms & Conditions</u> and the <u>Ethical guidelines</u> still apply. In no event shall the Royal Society of Chemistry be held responsible for any errors or omissions in this Accepted Manuscript or any consequences arising from the use of any information it contains.



Conductive	Hydrogel-based	Epidermal	Electrodes	for	Electrophysiological
------------	----------------	------------------	------------	-----	----------------------

^	B /	• 4	•	
<i>)</i>	VIO	nita	ring	5
_	TATO	11100	11115	٠

- 3 Jiawei Yang^{a,b}, Yi Liu^{a,b}, Wenqing Yan^{a,b}, Pengcheng Zhou^{a,b}, Zonglei Wang^{a,b}, Yuli
- 4 Wang^{a,b}, Yujie Zhang^{a,b}, Zongman Zhang^{a,b}, Fan Mo^c, Zichong Ji^{a,b}, Hossam Haick^b,
- 5 $Yan Wang^{a,b,d*}$

1

- 7 a Department of Chemical Engineering, Guangdong Technion-Israel Institute of
- 8 Technology 241 Daxue Road, Shantou, Guangdong 515063, China
- 9 b The Wolfson Department of Chemical Engineering, Technion-Israel Institute of
- 10 Technology, Haifa 3200003, Israel
- 11 ° Department of Materials Science and Engineering, Technion-Israel Institute of
- 12 Technology, Haifa 3200003, Israelpp
- d Guangdong Provincial Key Laboratory of Materials and Technologies for Energy
- 14 Conversion, Guangdong Technion-Israel Institute of Technology, 241 Daxue Road,
- 15 Shantou, Guangdong 515063, China

16

- 17 **Keywords**: Conductive hydrogels; epidermal electrodes; electrophysiological
- 18 monitoring

19

20 *E-mail: wang.yan@technion.ac.il; yan.wang@gtiit.edu.cn

View Article Online DOI: 10.1039/D5TC01896J

Abstract

21

22

23

24

25

26

27

28

29

30

31

32

33

34

35

36

Electrophysiological signals generated by human physiological processes offer critical insights for health monitoring and disease diagnosis, with their precise acquisition depending on high-performance electrodes. Conductive hydrogel-based epidermal electrodes, owing to their superior properties, demonstrate significant promise in electrophysiological monitoring. This review presents a comprehensive summary of the recent progress in the design and application of conductive hydrogels for epidermal electrophysiological electrodes. It first categorizes the various types of conductive hydrogel materials, highlighting recent advancements and their unique advantages as electrode interfaces. Subsequently, the key properties of conductive hydrogel-based epidermal electrodes are discussed, including conductivity, adhesion, stretchability, and gas-permeability. Then, state-of-the-art applications multiple across electrophysiological domains are introduced, ranging from electrocardiography, electromyography, electrooculogram, electroencephalography. and Finally, conclusion and future directions for the conductive hydrogel-based epidermal electrodes in electrophysiological monitoring are provided.

This article is licensed under a Creative Commons Attribution-NonCommercial 3.0 Unported Licence

Open Access Article. Published on 05 2025. Downloaded on 29/10/2025 01:38:06.

37

38

39

40

41

42

43

44

45

46

47

48

49

50

51

52

53

54

55

56

57

58

59

60

61

62

63

1. Introduction

Electrophysiological signals, including electrocardiogram (ECG), electromyogram (EMG), electroencephalogram (EEG), and electrooculogram (EOG), reflect critical human physiological processes, capturing bioelectric activity from heartbeats, muscle contractions, and neural functions¹. These signals are essential for diagnosing and monitoring cardiovascular conditions^{2, 3}, investigating neurological disorders^{4, 5}, supporting motor rehabilitation^{6, 7}, and advancing human-computer interaction systems^{8,9}. Characterized by high temporal and spatial complexity, these signals exhibit amplitudes ranging from microvolts to millivolts and frequencies spanning sub-hertz to hundreds of hertz¹⁰. Consequently, skin-mountable electrodes, as the cornerstone of electrophysiological signal acquisition, must combine high sensitivity, mechanical stability, stretchability, and conformal skin contact to maintain signal fidelity and improve clinical outcomes¹¹⁻¹⁵. Electrodes are generally categorized as invasive or non-invasive and serve as critical interfaces in biochemical sensing and human health monitoring, enabling efficient transmission and transduction between biological signals and electronic systems 16-20. Invasive electrodes, implanted to directly interface with target tissues, yield highresolution signals but are constrained by surgical risks and invasiveness, limiting their use in routine monitoring. Non-invasive electrodes, which capture signals through skin contact, are further divided into dry and wet electrodes. Dry electrodes, typically constructed from metals or rigid conductive materials, are valued for their reusability and ease of application but exhibit high stiffness, making it challenging to maintain intimate skin contact during dynamic deformation or movement²¹. This often results in skin-electrode contact gaps, elevating interface impedance and diminishing the signalto-noise ratio (SNR). Wet electrodes, such as commercial Ag/AgCl gel electrodes, utilize conductive gel to enhance skin-electrode contact for lower skin-electrode impedance, delivering high-quality signals in the short term²². However, they typically

65

66

67

68

69

70

71

72

73

74

75

76

77

78

79

80

81

82

83

84

85

86

87

88

89

90

91

rely on semi-liquid conductive gels, prolonged use may lead to discomfort, Paller 1896J reactions, or signal attenuation due to gel desiccation or skin irritation. Moreover, their rigidity hampers signal acquisition in dynamic settings. In recent years, advancements in flexible electronics have positioned epidermal electrodes made from flexible materials as compelling alternatives to conventional electrodes²³⁻²⁵. With exceptional stretchability (exceeding 200% strain), skin-matched low modulus, and ultra-thin profiles, these electrodes establish soft, stable skin interfaces, markedly reducing skin contact impedance and enhancing SNR. Consequently, the development of flexible epidermal electrodes optimized for long-term, stable signal acquisition has emerged as a pivotal research priority. Hydrogels are hydrophilic, cross-linked polymer networks that stably retain at least 10 wt % water within their three-dimensional structure²⁶. This structure imparts exceptional biocompatibility and a low Young's modulus, rendering hydrogels ideal for skin-contact applications^{27, 28}. Traditional hydrogels generally exhibit poor electrical conductivity, which impairs their ability to respond to external signals and restricts their effectiveness in electrophysiological signal acquisition. To address this, researchers have engineered conductive hydrogels by integrating conductive elements—such as conductive polymers, nanomaterials, or ions—achieving remarkable electrical performance²⁹⁻³⁵. Fine-tuning the proportions of these components enables precise control over hydrogel conductivity. Furthermore, through innovative structural and chemical designs, conductive hydrogels can exhibit superior mechanical toughness, high conductivity, self-adhesion, and breathability^{36, 37}. In recent years, conductive hydrogels have attracted significant research interest due to their promising applications in bioelectronics and wearable devices³⁸⁻⁴⁰. For example, Mo et al.⁴¹ reviewed recent advancements in ionic conductive hydrogels for skin sensor applications. While previous reviews have focused on specific subtypes—such as ionic liquid (IL)-based or nanomaterial-based conductive hydrogels—there remains a lack of comprehensive discussion on their role as epidermal electrodes for electrophysiological

monitoring^{25, 42-44}. Ding et al.⁴⁵ summarized the progress of conductive hydrogets 147/DSTC018963 electrophysiological signal acquisition; however, a holistic review encompassing material design, performance requirements, and practical applications in electrophysiological monitoring is still lacking. Therefore, this review aims to provide a comprehensive overview of the latest research trends in conductive hydrogel-based epidermal electrodes for electrophysiological applications. We begin by systematically introducing the typical materials used in conductive hydrogels, including design strategies for electronically conductive, ionically conductive, and hybrid conductive systems. Next, we discuss the key performance requirements for conductive hydrogel epidermal electrodes, such as conductivity, adhesion, stretchability, and gaspermeability. Finally, we summarize recent advances in their application for monitoring various electrophysiological signals (Fig. 1).

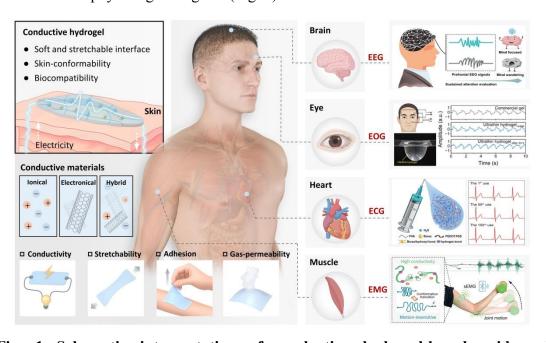


Fig. 1 Schematic interpretation of conductive hydrogel-based epidermal electrodes for electrophysiological monitoring. The left side illustrates representative types of conductive hydrogel materials, including ionic-based, electronic-based, and hybrid-based, along with their key properties, such as electrical conductivity, stretchability, adhesion, and gas permeability. The right side exhibits representative examples of electrophysiology monitoring, including ECG, EMG, EEG, and EOG. Reproduced with permission [46,47]. Copyright 2023, 2025, Wiley-VCH. Reproduced with permission [48]. Copyright 2024, Springer Nature. Reproduced with permission [49]. Copyright 2024, American Association for the Advancement of Science.

View Article Online

114

115

116

117

118

119

120

121

122

123

124

125

126

127

128

129

130

131

132

133

134

135

136

137

138

139

DOI: 10.1039/D5TC01896J

2. Materials of Conductive Hydrogel-based Epidermal Electrodes

Conductive hydrogels are composed of selectively introduced conductive fillers embedded in a cross-linked hydrophilic polymer matrix. Based on this fabrication method and configuration, we divide the epidermal electrodes based on conductive hydrogels into three categories: ionically conductive materials, electronically conductive materials, and hybrid conductive systems. In this section, we will discuss the representative materials of these three conductive hydrogel-based epidermal electrodes.

2.1 Ionically conductive hydrogel-based epidermal electrodes

Hydrogels, composed of one or more hydrophilic polymers, can absorb and retain significant amounts of water within their three-dimensional network structure⁵⁰. Leveraging the intrinsic hydrophilicity of hydrogel networks and the high mobility of ions in aqueous environments, ionically conductive hydrogels are typically formed by incorporating mobile ions into a crosslinked, water-rich polymer matrix. These hydrogels conduct electricity through the transport of ions such as Na⁺, K⁺, and Cl⁻ within the hydrated phase, which serves as the primary conduction medium. Under an applied electric field, the ions migrate through the network, enabling efficient ionic conduction⁵¹. Ion-conductive hydrogels are classified into three typical conductive mechanisms according to the migration mode of charge carriers and the characteristics of the conductive path: electrolyte-based, polyelectrolyte-based, and IL-based⁵². In electrolyte-based hydrogels, ionic conductivity results from the migration of small, dissociated ions (e.g., Na⁺, Cl⁻) through the water-rich polymer network. These ions introduced by dissolving inorganic salts or acids—can migrate freely within the hydrated phase when an electric field is applied. Electrolyte-based hydrogels are typically formulated by embedding these electrolytes into the polymer matrix, with commonly used examples including sodium chloride (NaCl), potassium chloride (KCl),

141

142

143

144

145

146

147

148

149

150

151

152

153

154

155

156

157

158

159

160

161

162

163

164

165

166

167

Open Access Article. Published on 05 2025. Downloaded on 29/10/2025 01:38:06.

BY-NG

This article is licensed under a Creative Commons Attribution-NonCommercial 3.0 Unported Licence.

lithium chloride (LiCl), sodium sulfate (Na₂SO₄), potassium dihydrogen phosphate^{9/D5TC01896J} (KH₂PO₄), tetramethylammonium chloride (TMACl), lactic acid, and various citrate salts⁴¹. Among typical chlorides, LiCl as a dopant can impart high conductivity to hydrogels, which significantly reduces skin contact impedance and enables the acquisition of highquality biopotential signals. Meanwhile, the introduction of ions can enhance the water retention of hydrogels, which is crucial for achieving long-term stable monitoring of biopotential signals⁵³. A typical example is that Li et al.⁵⁴ designed a body temperaturetriggered adhesive ionic conductive hydrogel based on biocompatible polyacrylamide (PAM), gelatin, LiCl, and sodium alginate (SA) (PGS hydrogel). Li⁺ can freely shuttle within the network structure of the PGS hydrogel, and the flow of these ions imparts conductivity to the hydrogel (Fig. 2a). The conductivity of the PGS hydrogel significantly increases with increasing LiCl content, rising from 0.28 ± 0.01 to $5.69 \pm$ 0.20 S/m (Fig. 2b). Furthermore, the conductivity of the PGS hydrogel remains largely stable under various stretching conditions (Fig. 2c). Even when the PGS hydrogel is stretched by 20% strain, the brightness of a light-emitting diode connected to it remains nearly unchanged. This demonstrates the stable conductivity of the PGS hydrogel in complex environments, facilitating high-quality electrophysiological monitoring. Polyelectrolytes serving as ionically conductive fillers bear covalently bound ionic groups (e.g., -COO⁻, -SO₃⁻) along the polymer backbone. In polyelectrolyte-based hydrogels, conductivity arises primarily from the migration of mobile counter-ions (e.g., Na⁺, K⁺) via ion hopping and segmental motion within the hydrated network^{55, 56}. These hydrogels offer good mechanical strength, especially in double-network structures, and their conductivity can be tuned by adjusting polymer concentration, crosslinking density, and counterion type, making them suitable for hydrated and physiological conditions. Typical polyelectrolyte fillers in hydrogels include polyacrylic acid (PAA) and its derivatives, chitosan, SA, etc⁵². Lu et al⁵⁷. developed a double-network (DN) polyelectrolyte hydrogel, integrating polymer chain entanglement, chemical

Open Access Article. Published on 05 2025. Downloaded on 29/10/2025 01:38:06.

This article is licensed under a Creative Commons Attribution-NonCommercial 3.0 Unported Licence.

168

169

170

171

172

173

174

175

176

177

178

179

180

181

182

183

184

185

186

187

188

189

190

191

192

193

194

195

crosslinking, and multiple strong and weak intermolecular interactions. The livedfore P/D5TC01896J consists of a PAM network and a polyelectrolyte network composed of polyelectrolytes (poly(diallyldimethylammonium and poly(diallyldimethylammonium chloride) chloride) (Fig. 2d). The mechanical properties and adhesive strength of polyelectrolyte DN hydrogels can be customized by modulating the proportions of PAM, polyelectrolyte, and cosolvent. The optimal formulation yields a tensile modulus of 10.8 kPa, a tensile strain at break of 1000% strain, and an adhesive strength of 37.8 kPa. Furthermore, the stability of the crosslinked PAM network, combined with the unique properties of polyelectrolytes that induce phase separation, ensures the hydrogel's stability even in salt solutions while exhibiting solvent-tunable transparency (Fig. 2e). Owing to the presence of polyelectrolytes, the hydrogel exhibits excellent conductivity. This conductive hydrogel can be stretched up to 1100% strain before reaching its breaking point while maintaining stable conductivity even under high tensile strain (Fig. 2f). hydrogels incorporate room-temperature ILs—such as IL-based 1-ethyl-3methylimidazolium bis(trifluoromethanesulfonyl)imide (EMIM-TFSI)—either as cosolvents or as dispersed conductive media within the hydrogel network. Their ionic conductivity stems from the unrestricted movement of both organic cations and anions, which act as charge carriers. These hydrogels exhibit high and humidity-independent ionic conductivity, excellent electrochemical stability, and strong anti-freezing and anti-drying properties. They are particularly suitable for long-term use in harsh environments and maintain good flexibility and thermal stability^{58, 59}. By polymerizing IL monomers into PIL, the inherent properties of IL can be transferred to the polymer chain, thereby obtaining ionic conductive hydrogels with high conductivity. Zhao et al. 60 prepared an ionic gel through one-step photoinitiated polymerization of 2,2,2trifluoroethyl acrylate and N-isopropylacrylamide (NIPAm) in the hydrophobic ILs 1ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide ([EMIm][TFSI]) (Fig. 2g). The ionic gel demonstrates exceptional transparency (94.8%), underwater self-

healing capability (up to 96%), toughness (3.93 MJ/m³), and

View Article Online DOI: 10.1039/D5TC01896J

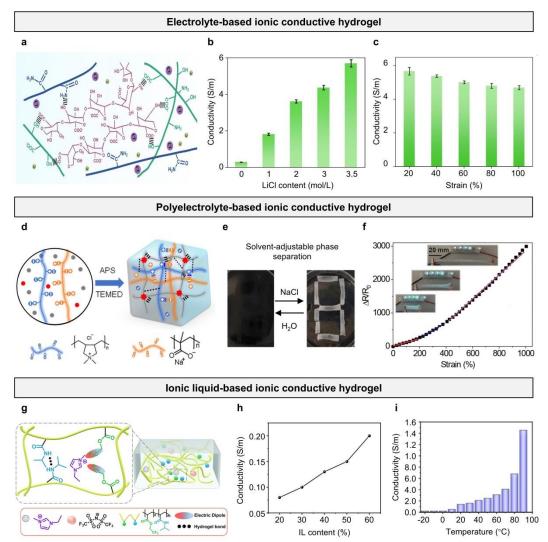


Fig. 2 Ionically conductive hydrogel-based epidermal electrodes. (a) PGS conductive hydrogel and its network structure. (b) Conductivity of PGS hydrogels with different LiCl contents. (c) Conductivity of PGS hydrogel under different strain. Reproduced with permission [54]. Copyright 2024, Elsevier. (d) Schematic diagram of the polyelectrolyte hydrogel preparation and structure. (e) The photograph illustrates the solvent-tunable transparency of the hydrogel electrolyte under induced phase separation. (f) Tensile strain induced relative resistance changes of the hydrogel. Reproduced with permission [57]. Copyright 2025, Royal Society of Chemistry. (g) Schematic diagram of the structure of the ionogel. (h) Conductivity of ionogel with different IL contents. (i) The ionic conductivities of the iongel in the temperature range from −20 to 90 °C. Reproduced with permission [60]. Copyright 2023, American Chemical Society.

underwater adhesion strength (102.77 ± 2.4 kPa). As the ILs mass percentage increased from 20% to 60%, the ionic gel's conductivity surged from 8.15 × 10⁻⁴ to 2.04 × 10⁻³

213

214

215

216

217

218

219

220

221

222

223

224

225

226

227

228

229

230

231

232

233

234

235

236

237

238

S/cm (Fig. 2h). Inspired by the excellent temperature responsiveness of $\Phi_{S,0}$ the position temperature sensitivity of the ionic gel was further characterized. Across a wide temperature range of -20 to 90 °C, the ionic gel exhibited high ionic conductivity (2 × 10^{-4} to 1.45×10^{-2} S/cm). Due to increased ion mobility, the ionic conductivity sharply increased with rising temperature (Fig. 2i).

2.2 Electronically conductive hydrogel-based epidermal electrodes

In electronically conductive hydrogel-based epidermal electrodes, the hydrogel matrix provides a soft, biocompatible scaffold, while embedded conductive materials enable efficient electron transport. Electrical conduction is achieved by incorporating intrinsically conductive components into the hydrogel network. These materials form continuous pathways that support charge transport through electron hopping or bandlike conduction. The conductivity depends on factors such as filler dispersion and connectivity, polymer crystallinity, doping level, and interfacial interactions^{61, 62}. Electron conduction primarily encompasses three categories: i) conductive polymers, including poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS), polypyrrole (PPy), and polyaniline (PANi)^{63, 64}; ii) carbon filler-based nanomaterials, such as carbon nanotubes (CNTs) and graphene oxide (GO)^{65, 66}; and iii) metal-filler nanoparticles, such as silver nanoparticles (AgNPs)^{67, 68}. Incorporating these materials into hydrogels yields electron-conductive hydrogels with enhanced performance. A typical example is that Huang et al.69 utilized PEDOT:PSS to promote the selfpolymerization of zwitterionic [2-(methacryloyloxy)ethyl]dimethyl-(3-sulfopropyl) (SBMA), proposing an in-situ formed conductive hydrogel (PSP) (Fig. 3a). This hydrogel exhibits exceptional elasticity (elastic recovery rate > 96%), robust adhesion strength (6.5 kPa), biocompatibility, and intrinsic antibacterial properties. The gelation process generates minimal heat (< 5 °C), enabling in-situ formation on the skin. Furthermore, the hydrogel achieves intimate skin contact, creating a highly conformal interface (Fig. 3b). The PSP hydrogel exhibits high conductivity, attributed to its

polycrystalline ionic network and conductive PEDOT:PSS. As the SBMA PEONTEMP/D5TC01896J

increases, electronic conductivity of the PSP hydrogel decreases from 0.08 to 0.03 S/m,

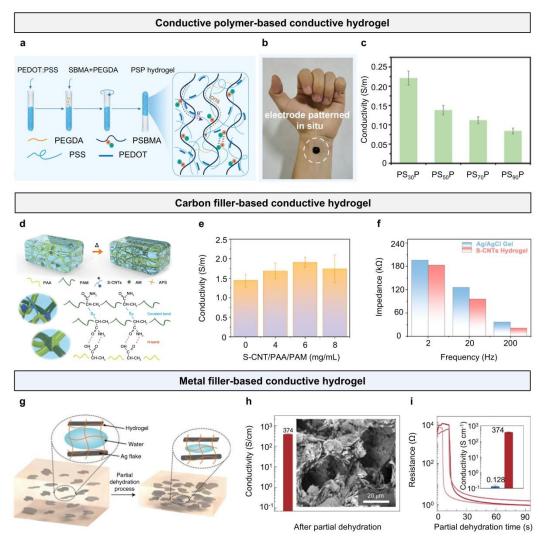


Fig. 3 Electronically conductive hydrogel-based epidermal electrodes. (a) Schematic diagram of the fabrication and promising applications of PSP hydrogel. (b) Photographs shows that the PSP hydrogel can be patterned on skin in situ. (c) The conductivity of the PSP hydrogel with different SBMA content. Reproduced with permission [69]. Copyright 2023, Wiley-VCH. (d) Schematic illustration of the preparation process of the S-CNTs/PAA/PAM hydrogel. (e) Conductivity of the prepared hydrogel with different S-CNTs content. (f) Impedance comparison at various frequencies of the S-CNTs/PAA/PAM hydrogel and commercial Ag/AgCl gel electrodes. Reproduced with permission [70]. Copyright 2025, American Chemical Society. (g) Composition of the conductive hydrogel composite composed of micrometre-scale Ag flakes and PAM-alginate hydrogel (Ag-hydrogel composite). (h) Conductivity of the Ag-hydrogel composite after the controlled partial dehydration process. (i) Conductivity of the Ag-hydrogel composite and micrographs of the composite after the controlled partial dehydration process. Reproduced with permission

View Article Online

DOI: 10.1039/D5TC01896J

257

258

259

260

261

278

279

280

281

282

283

284

256 [71]. Copyright 2021, Nature Publishing Group.

respectively. This reduction is attributed to the higher solid content and intensified inter- and intra-chain electrostatic interactions, which impede the formation of efficient conductive pathways. Nevertheless, the PSP hydrogel retains high conductivity, fully meeting the requirements for EMG monitoring (Fig. 3c).

Dai et al.⁷⁰ developed a hydrogel-based epidermal electrode composed of graphene

nanoplates, PAA, and PAM (S-CNTs/PAA/PAM). S-CNTs, enriched with graphene 262 263 nanoplates, are synthesized via a two-step sulphuration process employing thiourea and dibenzyl disulfide (Fig. 3d). The incorporation of sulfur atoms strengthens the 264 265 interfacial interactions between S-CNTs and PAA/PAM through C-S covalent bonding, significantly improving the hydrogel's mechanical performance (> 1200% strain) and 266 267 electrical conductivity (1.9 S/m). The incorporation of S-CNTs enhances the 268 conductivity of PAA/PAM hydrogels to a certain extent. The 6S-CNTs/PAA/PAM 269 hydrogel exhibits a conductivity of 1.9 S/m, surpassing that of the pristine PAA/PAM 270 hydrogel (1.44 S/m). The unique chain structure of S-CNTs on the polymer chains 271 shortens the charge migration pathway, thereby increasing the migration rate and 272 improving conductivity. However, excessive addition of S-CNTs leads to them 273 aggregation, which increases the overall resistance of the hydrogel and results in 274 reduced conductivity (Fig. 3e). Moreover, the S-CNTs/PAA/PAM hydrogel, employed 275 as an epidermal electrode for electrophysiological signal acquisition, demonstrates 276 significantly lower skin-contact impedance than commercial Ag/AgCl gel electrodes 277 across the 1–10⁵ Hz range (Fig. 3f).

Yunsik et al.⁷¹ designed a highly conductive, flexible hydrogel-based epidermal electrode by embedding micron-scale Ag particles within a PAM-alginate hydrogel matrix. A critical step in achieving high conductivity involves partial dehydration of the hydrogel matrix, facilitating the formation of percolation pathways by Ag flakes. Prior to partial dehydration, the Ag-hydrogel composite exhibits ionic conductivity with a low conductivity of ~0.13 S/cm. At this stage of the fabrication process, the volume fraction of Ag fillers (5 vol%) is insufficient to achieve percolation.

286

287

288

289

290

291

292

293

294

295

296

297

298

299

300

301

302

303

304

305

306

307

308

309

310

311

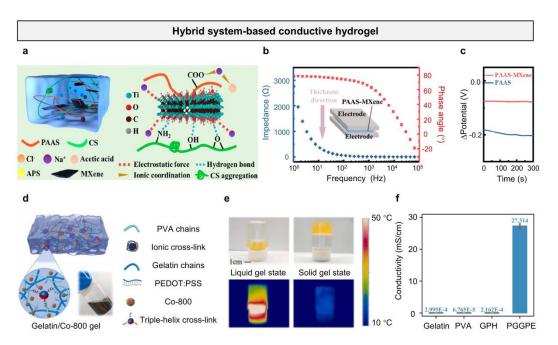
Subsequently, the conductivity of the composite increases significantly from 0.12 Ω 705 TC018963 374 S/cm following dehydration (Fig. 3h). During the partial dehydration process, Aghydrogel composites with varying amounts of Ag added in-situ yielded the resistance values of the hydrogel. The resistance of the Ag-hydrogel composite decreased exponentially after drying for 10–15 minutes, stabilizing at 1.14 \pm 0.35 Ω after 90 minutes, with a corresponding volume conductivity of 374 \pm 30.8 S/cm (Fig. 3i).

2.3 Hybrid conductive hydrogel-based epidermal electrodes

Hybrid conductive hydrogels combine ionic and electronic conductive components within a single network, effectively addressing the limitations of pure systems. Ionically conductive hydrogels offer good biocompatibility and flexibility but often exhibit reduced conductivity under low humidity or freezing conditions. In contrast, electronically conductive hydrogels provide stable and high conductivity but may compromise stretchability and lack sufficient ionic transport. By integrating both mechanisms, hybrid hydrogels form synergistic, bicontinuous ion-electron conduction networks that enhance charge transport kinetics and conductivity stability. This dualconduction design enables the hydrogel to maintain performance under mechanical deformation, temperature variation, and environmental stress. Additionally, hybrid networks allow tunable physical and electrical properties through molecular and structural engineering, such as tailoring polymer composition, conductive filler distribution, or crosslinking strategies⁷². For example, Luo et al. ⁷³ designed an MXeneinduced crosslinking fast-gelling hybrid conductive hydrogel electrode (PAAS) composed of acrylamide, NaCl, and MXene (Fig. 4a). Under the initiation of ammonium persulfate (APS), the C=C bonds of sodium acrylate are opened for polymerization, and then MXene acts as a crosslinker to rapidly form polymer chains. Meanwhile, the introduction of MXene increases the hydrogel's conductivity (2.3 S/m). In this system, sodium ions from NaCl and poly (sodium acrylate) impart ionic conductivity to hydrogel, while also establishing electrostatic interactions with MXene

View Article Online

and forming ionic coordination with acetic acid used for dissolving chitosan. Due to the /DSTC018963 addition of negatively charged MXene, the directional alignment of positively charged chitosan and sodium ions under an external electric field is restricted, reducing the hydrogel's polarization potential (change rate less than 6.5×10^{-4} V/min) (Fig. 4b and c).



317318

319

320

321322

323

324

325

326

327

328

329

330

331

332

333

312

313

314

315

316

Fig. 4 Hybrid conductive-based conductive hydrogel. (a) Composition of PAAS-MXene cross-linked by single-layer MXene. (b) Electrical impedance of Bode plot in the thickness direction of PAAS-MXene hydrogel. (c) Comparison of polarization potential values and polarization potential change values of PAAS and PAAS-MXene. Reproduced with permission [73]. Copyright 2022, American Chemical Society. (d) Schematic illustration of the multifunctional bio-hydrogel. (e) Conductivity of gelatin, PVA, GPH, and PEDOT:PSS/graphene/gelatin/PVA hydrogel. (f) Comparison of single heartbeat waveform recorded by hydrogel electrode and commercial electrode. Reproduced with permission [74]. Copyright 2023, Wiley-VCH. Wang et al. 75 developed a hybrid conductive hydrogel leveraging the synergistic effects of PEDOT:PSS/graphene and Na⁺ ions (Fig. 4d). The hydrogel (GPH), composed of a dual-crosslinked network of gelatin and polyvinyl alcohol (PVA), exhibits a unique thermally responsive reversible phase transition between a flowable fluid state and a viscoelastic gel state (Fig. 4e). Compared to pristine hydrogels (6.76 × 10⁻⁵ mS/cm), the conjugated structure of PEDOT:PSS in the hybrid conductive hydrogel facilitates electron mobility, while graphene sheets form a conductive network that supports

335

336

337

338

339

340

341

342

343

344

345

346

efficient electron and ion transport. This results in significantly enhanced effective phostco18963 conductivity, reaching 27.31 S/cm (Fig. 4f).

3. Properties of Conductive Hydrogel-based Epidermal Electrodes

To fulfill the essential requirements for electrophysiological signal monitoring, conductive hydrogel-based epidermal electrodes can be engineered with tailored properties, including conductivity, stretchability, and adhesion, using diverse synthesis and fabrication techniques (Table 1). This section outlines the essential properties required for conductive hydrogel epidermal electrodes, highlighting their critical role in ensuring the efficacy and reliability of electrophysiological monitoring.

Table 1 Summary of conductive hydrogel-based epidermal electrodes from different conductive mechanisms.

						1		
Conducti ve types	Hydrogel matrix	Conductivity materials	Conductivity (Test method)	Stretchability (%)	Adhesion (kPa)	Biocompatibility	Electrophysio logical signals	Ref.
	AM/BIS/Water	NaCl	1.3 S/m (EIS)	1326	14	-	ECG/EEG	76
	AAM/PEGDA/Water	LiCl	0.01 S/m (EIS)	2500	-	-	EOG/EEG	77
	PVA/b-PEI/Water	CaCl ₂	3.09 S/m (EIS)	1291	10	-	ECG/EMG/ EEG	78
	EGDMA/Water	VBIm-NTf ₂	0.0065 S/m (AC Resistance)	200	~400	-	ECG	79
	HEMA/SBMA/MBA/Wat er	LiTFSI	3 S/m (Four-point Probe)	287	7.5	High	ECG/EMG	80
Ionic	PAM/PDDA/Water	PMAANa	-	1000	19.2	-	ECG/EMG	57
Tonic	DMAEA/MBA/KPS/Wat er	ZM	-	1880	400	High	ECG/EMG	81
	AAm/LMA/KPS/Water		-	1410	-	High	ECG/EMG	82
	PVA/PAM/F-SiO ₂ /Water	CaCl ₂	10.58 S/m (EIS)	1450	26	-	ECG/EMG	83
	Gelatin glycerol/Borax/Water	Na ₂ SO ₄	9.3 S/m (EIS)	696	176	-	ECG/EMG/EE G/EOG	49
	Proanthocyanins/Guar gum/CNF/Water	FeCl ₃	0.023 S/m (Four-point Probe)	-	7.9	-	ECG/EMG	84
	EG/AAm/MBA/Water	DES	1.6 S/m (AC Resistance)	1660	0.01	High	ECG/EMG	85
	PVA/PDA/Water	PEDOT:PSS	2.18 S/m (Four-point Probe)	535	197	-	EMG	86
	PDA/Water	PEDOT:PSS	4 S/m (AC Resistance)	1300	22.4	High	ECG/EMG	87
,	PVA/SA/APS/FeCl ₃ /Wate r	PEDOT:PSS	0.256 S/m (EIS)	334	-	-	ECG/EMG/EE G	88
	NIPAm/HEA/Water	poly(Cu- arylacetylide)	3.1 S/m (EIS)	108	43	High	ECG/EMG	89
E1 .	APS/SA/MBAA/KCI/ CaSO ₄ /Water	PEDOT:PSS	-	50	64	High	EEG	90
Electroni c	PAM/AM/Silk fibroin/Water	MXene	0.25 S/m (AC Resistance)	1560	29	-	EMG/EOG	91
	TA/ACC/Water	LM	0.02 S/m (EIS)	1000	28.9	High	ECG/EMG	92
	PVA/Borax/Water	PEDOT:PSS	0.002 S/m (Two-point Probe)	10000	15.7	-	ECG/EMG	93
	PAA/AlNO ₃ /Water	MXene	0.7 S/m	2400	494.2		ECG/EMG	94
	PAA/PVA/Water	RGO	0.11 S/m (Two-point Probe)	-	5.4	High	ECG/EMG/E OG	95
	PAM/Alginate/Water	Ag	37400 S/m (Four-point Probe)	250	-	-	EMG	71
Hybrid	PEGDA/Water	PEDOT:PSS /SBMA	0.03 S/m (EIS and DC Resistance)	390	6.5	High	ECG/EMG	69
	PAA/SA/AD/ADQ/Water	MXene/LiCl	1.8 S/m (EIS)	800	~27	High	ECG	96
	HA/Water/Glycerin	GNs/KCl/Na Cl	0.33 S/m (EIS)	-	~5.5	High	EEG	97
	PEDOT:PSS/Fe/Water	PEDOT:PSS/ Fe	1.25 S/m (EIS)	15	-	High	ECG	98
	PAA/TOCNFs/Water	PEDOT:PSS/ Al(TFSI) ₃	7.1 S/m (EIS)	770	28	High	ECG/EMG	99
	PVA/Water	PPy/FeCl ₃	80 S/cm (Four-point Probe)	36	-	High	EMG	100

BIS: N,N '-Methylenebisacrylamide; PEGDA: poly(ethylene glycol) diacrylate; HEMA: 2-

Hydroxythyl methacrylate; SBMA: [2-(Methacryloxy) ethyl] dimethyl-(3-sulfo-propyl) ammonium

View Article Online

347

348

349

350

351

352

353

354

355

356

357

358

359

360

361

362

363

364

365

366

367

368

369

370

371

372

373

hydroxide; MBA: N,N '-Methylenebisacrylamide; SBMA: zwitterionic 12-10.1039/D5TC018965 (methacryloyloxy)ethyl]dimethyl-(3-sulfopropyl); LiTFSI: bistrifluoromethanesulfonimide lithium salt; PDA: polydopamine; NIPAm: N-isopropylacrylamide; HEA: 2-hydroxyethyl acrylate; GNs: graphite nanoparticles; HA: hyaluronic acid; DMAEA: 2-(Dimethylamino) ethyl acrylate; MBA: N,N'-methylenebis(acrylamide); KPS: potassium persulfate; LMA: lauryl methacrylate; ACC: calcium carbonate. EIS: electrochemical impedance spectroscopy.

3.1 Conductivity

The conductivity of hydrogel-based epidermal electrodes significantly influences the performance of bioelectronic interface electrodes, crucial for enabling efficient electrical signal transmission between biological tissues and electronic components while ensuring stable electrophysiological communication¹⁰¹. The polymer network serves as a structural scaffold, while the conductive fillers confer electrical conductivity to the hydrogel^{32, 102}. To date, a range of conductive materials, encompassing both electronic and ionic conductive fillers, have been employed in the fabrication of conductive hydrogels for epidermal electrodes¹⁰². Ionic conductive fillers are incorporated into hydrogel systems by introducing acids (e.g., HCl, H₂O₄ H₃O₄, ionic compounds (e.g., LiCl, FeCl₃ AlCl₃, NaOH, KOH), or ionic liquids (e.g., 1-ethyl-3-methylimidazolium sulfate), which release free ions to significantly enhance electrical conductivity^{41, 103}. For instance, Zhang et al. ¹⁰⁴ prepared highly conductive hydrogels by adding KOH and NaOH to a carboxymethyl chitosan (CECT)/PAM-based hydrogel (CTA). The ionic conductivity of the hydrogel samples was measured using electrochemical impedance spectroscopy, with conductivity ranging from 0.38 to 0.62 S/m. Another way to achieve high conductive performance is typically to incorporate electronic conductive fillers (such as intrinsically conductive polymers, metal nanoparticles, or carbon-based nanomaterials) into hydrogels 105, 106. A typical example is that Wang et al¹⁰⁷, employed a synergistic approach combining freeze-drying and

375

376

377

378

379

380

381

382

383

384

385

386

387

388

389

390

391

392

393

394

395

396

397

398

399

400

401

Open Access Article. Published on 05 2025. Downloaded on 29/10/2025 01:38:06.

This article is licensed under a Creative Commons Attribution-NonCommercial 3.0 Unported Licence.

salting-out treatment to prepare a conductive hydrogel composite based of psilve p/D5TC01896J nanowires (AgNWs) and PVA (Fig. 5a). This method successfully constructed a layered hydrogel structure and significantly enhanced the local concentration of AgNWs by inducing continuous phase separation. The resulting conductive hydrogel composite exhibited remarkable properties, including ultra-high conductivity, excellent stretchability (480% strain), and outstanding biocompatibility. The phase separation of PVA in the hydrogel evolves with the extension of the salting-out time. By optimizing the salting-out treatment time, the researchers successfully controlled the phase separation process, resulting in a hydrogel with a conductivity exceeding 1739 S/cm (Fig. 5b). Due to the high concentration of AgNWs, the prepared hydrogel still maintains high conductivity under stretching, which is superior to other types of hydrogels (Fig. 5c). Zhang et al. 108 developed a liquid metal (LM)-doped PVA-LM hydrogel, incorporating LM microdroplets within a tannic acid (TA)-modified PVA matrix (Fig. 5d). LM microdroplets were employed as crosslinkers to enhance mechanical properties while providing exceptional conductivity, reaching a peak value of 217895 S/m. The sedimentation time of LM profoundly influences the hydrogel's conductivity, with the non-sedimented PVA-LM hydrogel exhibiting a significantly lower conductivity of 0.004 S/m. After a sedimentation time of 30 minutes, the conductivity of the PVA-LM hydrogel surged by five orders of magnitude to 455 S/m, reflecting significant LM microdroplet precipitation (Fig. 5e). The PVA-LM hydrogel was subjected to resistance measurements over 1200 stretching cycles (40% strain), during which its resistance progressively decreased from 4.8 to 4.5 Ω (Fig. 5f). This suggests mechanical sintering of LM microdroplets on the hydrogel surface under external forces, concurrently enhancing its conductivity. For epidermal electrodes, both ionic and electronic conductivity are essential yet functionally distinct. Ionic conductivity plays a crucial role in interfacing with the skin, which is a naturally ionic medium. Hydrogels with high ionic conductivity can form low-impedance, conformal contacts with the stratum corneum, facilitating effective

coupling with bioelectric signals such as ECG, EMG, or EEG. This enables stable, high postco18963 fidelity acquisition of weak biopotentials with reduced signal loss or distortion.

Conductivity

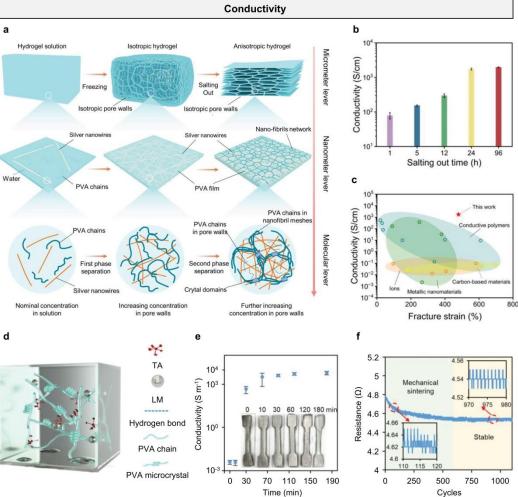


Fig. 5 Conductivity. (a) Schematic illustrating the fabrication procedure of AgNWs-PVA hydrogel composites by combining freezing and salting-out treatments. (b) Conductivity of the AgNWs-PVA hydrogel composites with varying durations of salting-out treatment. (c) Comparison of conductivity and fracture strain of the Ag-PVA hydrogel with other conductive hydrogels based on different conductive fillers. Reproduced with permission [107]. Copyright 2024, Wiley-VCH. (d) Schematic of PVA-LM hydrogel network. (e) Conductivity of PVA-LM hydrogels with different aging times. (f) The resistance response of PVA-LM hydrogel under cyclic stretching for 1000 cycles. Reproduced with permission [108]. Copyright 2024, Wiley-VCH. Conversely, electronic conductivity is vital for rapid and efficient transmission of the collected signals to external readout devices. Materials such as PEDOT:PSS, CNTs, or MXene can form continuous electron pathways that minimize resistance across the electrode structure. In hybrid conductive hydrogels, the synergy between these two

419

420

421

422

423

424

425

426

427

428

429

430

431

432

433

434

435

436

437

438

439

440

441

442

443

444

conduction mechanisms enables high SNR, reduced motion artifacts, and stable/D5TC018963 performance under varying environmental and mechanical conditions. By combining the skin-compatible interface of ionic systems with the robust signal transfer capability of electronic systems, hybrid hydrogels offer a balanced and integrated solution for long-term, reliable epidermal bioelectronic applications.

3.2 Adhesion

Superior adhesive properties enable hydrogel-based epidermal electrodes to effectively adhere to the skin, minimizing skin-contact impedance and enhancing the quality and stability of monitored signals¹⁰⁹. To enhance the adhesive properties of conductive hydrogel-based epidermal electrodes, there are three typical mechanisms to enhance the adhesiveness of conductive hydrogel epidermal electrodes: physical adhesion, chemical adhesion, and biomimetic strategies¹¹⁰. In physical adhesion, external stimuli, including pH variations¹¹¹, temperature fluctuations¹¹², ultraviolet light¹¹³, magnetic fields¹¹⁴, and electric fields¹¹⁵, can be utilized to modulate the adhesive properties of hydrogels. A typical example is that Liu et al. 116 proposed an electro-adhesion strategy with electrically programmable strength for universal and super-strong hydrogel bonding. The hydrogel is composed of PAA-Fe-Li. Owing to the robust hydration capacity of Li⁺ ions, they significantly modulate polymer chain interactions within the hydrogel, thereby enhancing its interfacial bonding through surface diffusion and accumulation (Fig. 6a). Driven solely by entropy related to the concentration gradient of the involved ions, PAA-Fe-Li exhibits effective adhesion to PVA hydrogels. After 5 seconds of contact, the adhesion strength and adhesion energy reach 0.24 MPa and 660 J/m², respectively. Subsequently, under a positive DC voltage, the adhesion efficiency of PAA-Fe-Li to PVA at ambient temperature significantly increased by ≈24 times (Fig. 6b). The peak adhesion strength and adhesion energy of PAA-Fe-Li with PVA substrates can reach 1.2 MPa and 3750 J/m², respectively. Adhered PAA-Fe-Li and PVA samples with a cross-sectional diameter of 6 mm can withstand at least 5 kg of

View Article Online

DOI: 10.1039/D5TC01896J

weight (Fig. 6c).

445

446

447

448

449

450

451

452

453

454

455

456

In chemical adhesion, the adhesive effect is mainly caused by chemical bonding and/or non-covalent interactions between reactive groups in the hydrogel and on the skin surface^{110, 117}. Covalent bonds play a predominant role in hydrogel adhesion due to their higher bond energy relative to other interactions¹¹⁸. Non-covalent interactions, including hydrogen and ionic bonds, are extensively employed in the fabrication of

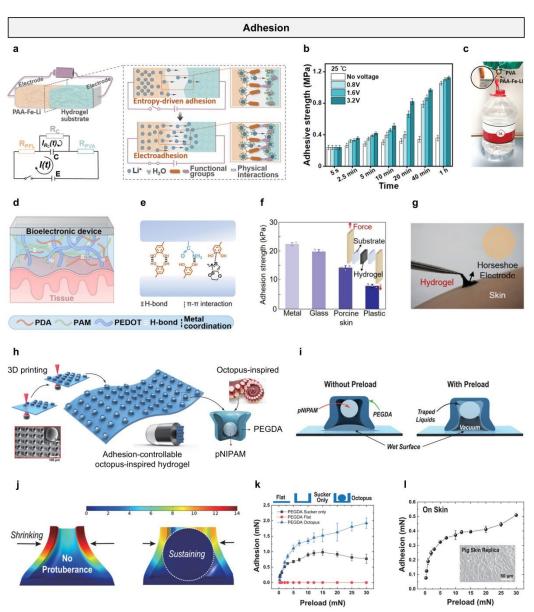


Fig. 6 Adhesion. (a) Mechanisms for strong interfacial bonding between the PAA-Fe-Li hydrogel and hydrogel substrate and schematic illustration of the proposed circuit model for electro-adhesion. (b) Adhesive strength of electro-adhesion between PAA-Fe-Li and PVA hydrogels at 25 °C. (c) Strong adhesion (> 5 kg) demonstration between PAA-Fe-Li and PVA. Reproduced with permission [116]. Copyright 2022, Wiley-VCH.

459

460

461

462

463

464 465

466 467

468 469

470

471

472

473

474

475

476

477

478

479

480

481

482

483

484

485

486

487

488

489

Open Access Article. Published on 05 2025. Downloaded on 29/10/2025 01:38:06.

BY-NG This article is licensed under a Creative Commons Attribution-NonCommercial 3.0 Unported Licence.

(d) Schematic illustration of a conventional bioelectronic device adhering directly to hotocolors of the conventional bioelectronic device adhering directly to hotocolors of the conventional bioelectronic device adhering directly to hotocolors of the conventional bioelectronic device adhering directly to hotocolors of the conventional bioelectronic device adhering directly to hotocolors of the conventional bioelectronic device adhering directly to hotocolors of the conventional bioelectronic device adhering directly to hotocolors of the conventional bioelectronic device adhering directly to hotocolors of the conventional bioelectronic device adhering directly to hotocolors of the conventional bioelectronic device adhering directly to hotocolors of the conventional bioelectronic device adhering directly to hotocolors of the conventional bioelectronic device adhering directly adhered to hotocolors of the conventional bioelectronic device adhered to hotocolors of tissue. (e) Noncovalent interactions among dopamine methacrylate, PEDOT, PDA, and PAM chains. (f) Adhesion strength of hydrogel to different substrates by lap shear test. (g) Image of a hydrogel bioelectronic device for EMG and ECG monitoring. Reproduced with permission [87]. Copyright 2025, American Chemical Society. (h) Fabrication process of a polyethylene glycol diacrylate hydrogel. (i) Schematic illustration with finite element method simulation showing the adhesion mechanism of the sucker architecture with and without the inner protuberance structure. (i) Representative time-dependent profiles of adhesion results for the hydrogel patterns and flat glass for comparison. (k) Adhesion forces for different preloads (0.3–30 mN) in the samples of the inner pNIPAM and outer pNIPAM. (1) Adhesion results for different preloads (0.3 to 30 mN) in underwater conditions measured on the pig skin replica shown in inset. Reproduced with permission [119]. Copyright 2022, Wiley-VCH. adhesive hydrogels to enhance interfacial adhesion¹²⁰. Hydrogen bonds, distinguished for their adjustable strength, biocompatibility, and reversible bonding capabilities, represent a key intermolecular force. Generally, individual hydrogen bonds in hydrogels are weak, necessitating their synergistic combination with other interactions or the formation of multiple hydrogen bonds to enhance adhesion¹²¹. For example, Lao et al.87 developed a hydrogel with inherent adhesion and conductivity by integrating PAM-polydopamine (PDA) into conductive PEDOT, exhibiting a tissue-mimetic modulus (Fig. 6d). The adhesion is formed through non-covalent interactions of hydrogen bonds and π - π interactions with polydiacetylene and PAM chains (Fig. 6e). The shear strength of the hydrogel, adhered to various substrates including metal, glass, pig skin, and plastic, was measured at 22.4, 19.8, 14.2, and 7.9 kPa, respectively (Fig. 6f). Due to its ultra-soft and strong adhesive properties, the conductive hydrogel epidermal electrode can form a tight and robust interface with human tissue, thereby reducing artifacts caused by body movement (Fig. 6g). In biomimetic structures, the adhesion mechanisms of various organisms (including tree frogs, mussels, octopuses, and geckos) have attracted numerous researchers and inspired the development of biomimetic hydrogels^{122, 123}. For instance, the eight arms of an octopus are equipped with numerous suckers, each generating negative pressure via muscle control to produce robust adhesion¹²⁴. This mechanism, independent of chemical adhesives,

enables secure attachment to wet or irregular surfaces, such as marine rocks of preys/DSTC018963

These attributes inspire the development of high-performance, reusable biomimetic adhesive materials tailored for complex environments. Inspired by the octopus's orifice,

Lee et al. 119 printed polyethylene glycol diacrylate (pNIPAM) hydrogel to form an outer wall, completing an internal dome-shaped structure mimicking octopus protrusions, which enhanced dry and wet adhesion (Fig. 6h). The pNIPAM achieves robust wet adhesion under external preload by inducing cohesive forces among liquid molecules at interacting wet surfaces (Fig. 6i and 6j). Due to the enhancement of suction, the sucker with an internal dome-like protrusion structure can enhance adhesion in wet environments (Fig. 6k). Adhesion was evaluated under preloads ranging from 0.5 to 30 mN on a skin-mimicking surface. Owing to the roughness of pig skin, adhesion on pig skin was approximately three times lower or more than that on a smooth glass surface (Fig. 6l).

3.3 Stretchability

Beyond essential conductivity and adhesion, superior tensile performance is a critical attribute that positions hydrogels as ideal materials for stretchable epidermal electrodes. This capability allows hydrogel-based electrodes to effectively accommodate body deformations while sustaining conformal contact, ensuring reliable signal transmission²⁷. Typical strategies for enhancing the mechanical properties of hydrogel epidermal electrodes include constructing multiple crosslinked network structures, adding nanocomposites, introducing supramolecular interactions, etc^{102, 125}. For example, Zhang et al¹²⁶. developed an all-cellulose hydrogel with exceptional stretchability by introducing supramolecular structures through the ring-opening reaction of cellulose's anhydroglucose units via dehydration (Fig. 7a). The hydrogel's tensile properties were enhanced by modifying the cellulose structure and tuning interchain interactions. To increase chain flexibility, periodate oxidation was employed to selectively cleave the C2–C3 bond, converting secondary hydroxyl groups into

518

519

520

521

522

523

524525

526

aldehyde groups and thereby opening the anhydroglucose ring structure. The resulting/D5TC01896J hydrogel exhibited a record-breaking tensile of 42200% strain. The mechanical properties of the hydrogel were significantly influenced by the content of diol cellulose nanorods (DCNRs). As the DCNR content increased from 23.3 to 28.8%, the tensile stress rose progressively from 7 to 25 kPa. When the DCNR content ranged from 23.3 to 28.8%, the strain of the hydrogels exceeded 10000% strain. (Fig. 7b). To further validate the hydrogel's stretchability, a 1 cm hydrogel was stretched to 4.42 m,

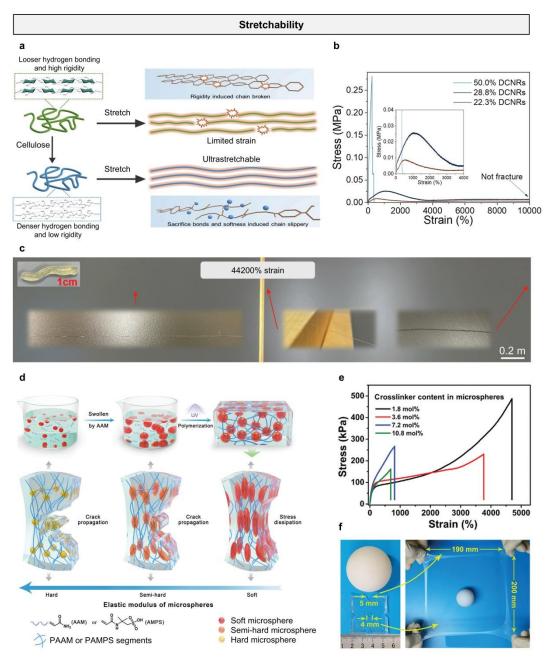


Fig. 7 Stretchability. (a) Schematic illustration of the cleavage of rigid anhydroglucose unit rings enhances hydrogel chain flexibility and facilitates the reformation of

529

530

531

532

533

534

535

536

537

538

539

540

541

542

543

544

545

546

547

548

549

550

551

552

553

554

555

556

557

hydrogen bonds during tensile deformation. (b) Stress-strain curves under different view Article Online DCNRs contents. (c) Photos of DCNRs hydrogel before and after stretching. Reproduced with permission [126]. Copyright 2024, Wiley-VCH. (d) Schematic illustration of the preparation process and mechanism of super-stretchable hydrogel. (e) Tensile curves of hydrogels under different cross-linker concentrations. (f) Tear resistance of 5 mm long hydrogel under pre-cut equilateral triangle notch. Reproduced with permission [127]. Copyright 2022, Wiley-VCH. corresponding to a strain of 42200% (Fig. 7c). Ji et al. 128 introduced an IL imidazolium salt with a urea backbone (UL) and incorporated sulfobetaine methacrylate (SBMA) and acrylamide (AM) into the hydrogel system (ULAS) through simple one-pot copolymerization. Supramolecular interactions within the IL strengthen the noncovalent bond network of hydrogel-based epidermal electrodes. As the IL concentration increases, the hydrogel exhibits markedly enhanced stress, achieving an elongation at break of 1075% strain and a fracture stress of 343 kPa. Constructing physically crosslinked, entangled, and micelle-crosslinked networks has been demonstrated as an effective way to enhance the mechanical properties of hydrogels. Li et al¹²⁷. prepared a highly stretchable hydrogel with an interpenetrating entangled network by using PAM as the monomer and poly(1-acrylamido-2-methylpropanesulfonic acid) (PAMPS) as the physical crosslinking network, synthesized via UV-initiated free radical polymerization (Fig. 7d). The mechanical properties of hydrogels can be tuned by varying the crosslinker concentration. The fracture strain of hydrogels decreases rapidly with increasing crosslinking degree, reaching 4700% strain at a crosslinker content of 1.8 mol% (Fig. 7e). Furthermore, the hydrogel demonstrates outstanding tear resistance. Hydrogels with a 5 mm pre-cut notch extended to 190 mm and 200 mm under tensile stress without crack propagation. The area of a central circular pre-cut notch expanded approximately 2700-fold during cyclic biaxial stretching (Fig. 7f). Li et al.⁵⁴ developed a hydrogel with a multiply crosslinked network by incorporating gelatin and SA into PAM. With the addition of SA, a more complex network structure is formed between PAM/gelatin and SA through hydrogen bonds and electrostatic interactions between molecular chains. As the SA content rises from 0 to 0.8 wt%, the hydrogel's tensile stress increases from 61.3 to 229.3 kPa, with the elongation at break improving from

559

560

561

562

563

564

565

566

567

568

569

570

571

572

573

574

575

576

577

578

579

580

581

582

583

584

View Article Online DOI: 10.1039/D5TC01896J

428.4 to 706.4% strain.

3.4 Gas-permeability

Water vapor and sweat accumulation at the skin-electrode interface can lead to skin irritation and epidermal electrode failure during prolonged monitoring¹²⁹. Employing gas-permeable conductive hydrogel-based epidermal electrodes has proven an effective solution to this challenge¹³⁰. Common strategies for enhancing the gas-permeability of conductive hydrogel-based epidermal electrodes include fabricating ultrathin and porous structures^{129, 131}. A typical example is that Zhang et al.⁴⁹ introduced an ultrathin polyurethane (PU) nanomesh-reinforced gas-permeable hydrogel. The hydrogel was fabricated by immersing the PU nanomeshes in a temperature-sensitive gelatin-based hydrogel solution (Fig. 8a). The resulting ultrathin hydrogel has a thickness of ~10.3 μm (Fig. 8b), with excellent mechanical strength (up to 696% strain) and high skin adhesion (area adhesion energy of 176.8 µJ/cm²). Owing to its ultrathin geometry and porous structure, the hydrogel demonstrates a water vapor transmission rate (WVTR) of 1669.3 ± 23.5 g/m²/day, closely comparable to that of an open bottle (1742.8 \pm 18.3 g/m²/day) (Fig. 8c). The resultant ultrathin hydrogel-based epidermal electrode enables long-term, continuous, high-precision electrophysiological monitoring for up to 8 days under everyday conditions. Cheng et al.¹³² developed a cold lamination-based approach to fabricate breathable hydrogel films with precisely controlled thickness and scalability. A mixed solution of PAM and SA is cast between two polyethylene terephthalate (PET) support films and then rapidly passed through the gap between two rollers. The prepared PET-wrapped hydrogel precursor film is cured under ultraviolet light to complete in-situ gelation (Fig. 8d). The prepared hydrogel film thickness can be controlled to a minimum of 7 µm. The ultrathin hydrogel film has good skin conformability, and cross-sectional SEM images show close contact between a fingerprint replica and a 10 µm thick hydrogel film (Fig. 8e). Due to its ultrathin characteristics, the hydrogel film has good

breathability, measured by transepidermal water loss (TEWL). A 50 µm thick hydrogeP/D5TC018963 film and a 3 µm thick impermeable parylene film were attached to the skin, respectively. After applying the ultrathin acrylic film for 2 hours, the initial skin hydration level increased from approximately 25 to 45%. Accompanying this, the TEWL significantly increased from ~7 to 12 g/m²/day. After removing the acrylic film, the skin moisture level dropped to ~28% within 30 minutes, and the TEWL also decreased (Fig. 8f).

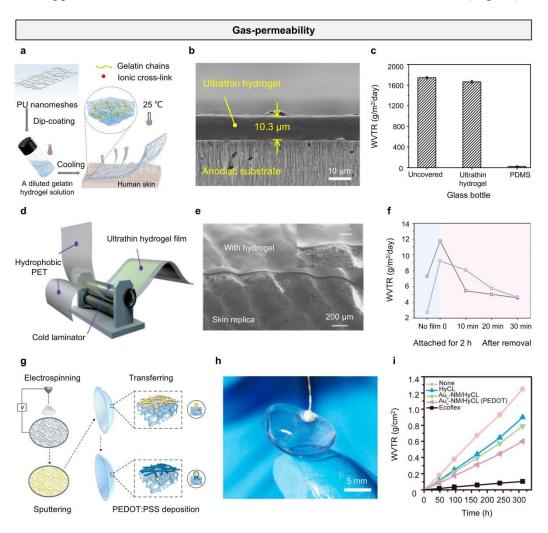


Fig. 8 Gas-permeability. (a) Schematic illustration of the design concept of PU nanomesh-reinforced hydrogels. (b) Cross-sectional SEM image of the ultrathin hydrogel attached on an anodisc substrate. (c) Comparison of WVTR for an uncovered bottle, a bottle covered with 1000-μm-thick PDMS film, and a bottle covered with ~10-μm-thick ultrathin hydrogel, respectively. Reproduced with permission [49]. Copyright 2024, American Association for the Advancement of Science. (d) Schematic of the cold-lamination method to produce large-area ultrathin hydrogel films. (e) SEM images of a 10 μm thick hydrogel film attached to the fingerprint replica. (f) TEWL and the skin hydration level test by attaching a 3 μm thick parylene film on the forearm of the

- volunteer. (g) Schematic illustration of the gas-permeable, irritation-free, transparenty of the paradyses o
- 602 hydrogel film. Reproduced with permission [132]. Copyright 2022, Wiley-VCH. (h)
- Picture of a contact lens-based eye interfacing device based on the hydrogel film. (i)
- WVTR of open bottle (none), pure hydrogel ocular contact lens (HyCL), HyCL with
- gold-coated nanofiber mesh (Auc-NM/HyCL), Auc-NM/HyCL with deposition of
- 606 PEDOT:PSS, and Ecoflex film as a function of elapsed time. Reproduced with
- 607 permission [133]. Copyright 2019, American Chemical Society.
- Wei et al.¹³³ reported a breathable, non-irritative, and transparent hydrogel film. The
- gas-permeable film was fabricated using a metal-coated nanofiber network (metal_c-NM)
- as the electronic conductor and a commercial hydrogel contact lens as the substrate,
- with in situ electrochemical deposition of PEDOT:PSS employing the metal_c-NM as
- the electrode (Fig. 8g). The resultant hydrogel film exhibits an optical transparency of
- 613 90% (Fig. 8h). Owing to the highly porous structure of the metal_c-NM film, the
- underlying hydrogel substrate retains its breathability, enabling the hydrogel film to
- demonstrate excellent breathability with a WVTR of approximately 69.2 mg/cm²/day,
- significantly exceeding that of an impermeable Ecoflex film at 7.98 mg/cm²/day (Fig.
- 617 8i).

619

626

4. Applications of Conductive Hydrogel-based Epidermal Electrodes in

Electrophysiological Monitoring

- 620 Conductive hydrogel-based epidermal electrodes, due to their good conductivity,
- outstanding skin adhesion and compliance, excellent stretchability, and superior
- breathability, have shown great potential in the field of electrophysiological signal
- 623 monitoring. In this section, we will summarize the representative progress of
- 624 conductive hydrogel-based epidermal electrodes in ECG, EMG, EEG, and EOG
- 625 monitoring.

4.1 Electrocardiogram monitoring

627 ECG signals play a pivotal role in the medical field, being extensively utilized for

629

630

631

632

633

634

635

636

637

638

639

640

641

642

643

644

645

646

647

648

649

650

651

652

653

654

655

diagnosing cardiovascular diseases 134-137, and assessing cardiac health 1380 1469 9/D5TC01896J Conductive hydrogel-based epidermal electrodes possess distinctive properties, including high conductivity, excellent stretchability, and strong interfacial adhesion¹⁴¹, ¹⁴². These attributes enable them to achieve superior SNR and sensitivity compared to traditional rigid electrodes, thereby enhancing the accuracy and reliability of monitoring and providing more precise data to support clinical diagnostics and treatment. A typical example is that Yang et al.¹⁴³ developed a wireless Nepenthes-inspired hydrogel (NIH) hybrid system. The hybrid system consists of a skin electrode assembled from a flexible circuit module, an electrode connector with a hollow structure, a stretchable silicone joint, and three Nepenthes-inspired hydrogel interface layers (Fig. 9a). The DN hydrogel based on PVA/PAA enhances its adhesion to the skin (3.9 kPa). To evaluate the practicality of the NIH hydrogel system during motion, the system was laminated on the skin above the subject's heart to collect ECG signals (Fig. 9b). To evaluate the stability of the NIH system, heart rate curves were recorded using the system and a commercial device (i.e., Polar H10 heart rate strap) under motion conditions, respectively. The results demonstrate that data from both systems reveal nearly identical heart rate variation trends, with a relative deviation of less than 2.6% over 5 minutes, validating the accuracy of the NIH system (Fig. 9c). Inspired by the pentaradial symmetry of starfish, Chen et al.¹⁴⁴ introduced a starfishlike wearable hydrogel-based bioelectronic system for high-fidelity ECG monitoring. The skin interface layer consists of conductive hydrogels for five sensing pads and nonconductive hydrogels for the central hub, while the serpentine arms remain independent. The device is constructed on a 25 µm-thick polyimide (PI) substrate, utilizing copper traces as conductive pathways. Its pentaradial design incorporates five serpentine arms, each equipped with an independent sensing element at its tip, all linked to a central electronic hub for data processing and wireless transmission. Signals are processed by a 32-bit microcontroller and transmitted wirelessly, with analyzed data presented

657

658

659

660

661

This article is licensed under a Creative Commons Attribution-NonCommercial 3.0 Unported Licence 662 Open Access Article. Published on 05 2025. Downloaded on 29/10/2025 01:38:06. 663 664 665 666

667

668

669

670 671

intuitively on a mobile device (Fig. 9d). By employing a conductive adhesive hwdrogep/D5TC01896J as the electrode-skin interface and a mechanically decoupled starfish-inspired device design (Fig. 9e), this device enables high-fidelity ECG recordings across diverse motion states, sustaining an SNR of approximately 35 dB even during running. The system enables real-time, high-precision diagnosis of cardiac conditions by integrating ECG, seismocardiogram, and gyrocardiogram signals with a deep learning model, achieving classification accuracies of 91.31% for normal conditions, 94.03% for atrial

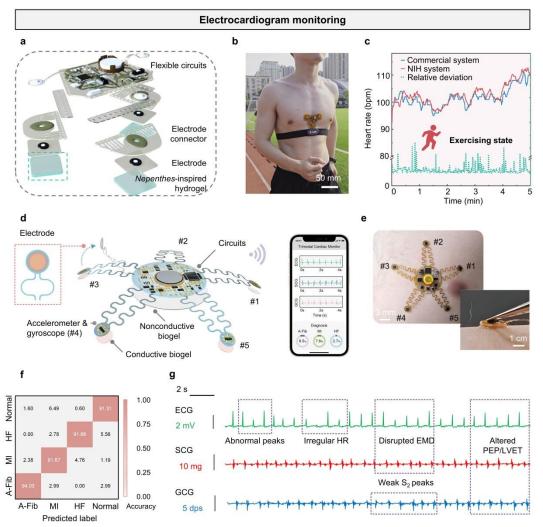


Fig. 9 Electrocardiogram monitoring. (a) Exploded 3D model of the NIH hybrid system. (b, c) Heart rate and relative deviation curves measured by the NIH system and commercial system under the subject's exercising states. Reproduced with permission [143]. Copyright 2024, Wiley-VCH. (d) Schematic illustration of the starfish-like device for trimodal cardiac monitoring during motion. (e) Optical image showing the device applied to the chest skin, with the five strategically positioned sensing pads, and optical image, showing robust adhesion of the sensing pad to human skin, facilitated by the conductive hydrogels. The image features a tweezer used to stretch the sensing pad

674

675

676

677

678

679

680

681

682

683

684

685

686

687

688

689

690

691

692

693

694

695

696

697

698

699

700

from the skin. (f) Confusion matrix, demonstrating the classification accuracy for the skin. (g) Confusion matrix, demonstrating the classification accuracy for the skin. predicting each type of heart disease and normal signals in the test set. During use, the starfish-like device can display the real-time classification probabilities on smart devices. (g) Cardiac electrical and mechanical signals collected by the starfish-like wearable device from a HF patient during walking; dps, degrees per second. Reproduced with permission [144]. Copyright 2025, American Association for the Advancement of Science. fibrillation, 91.67% for myocardial infarction, and 91.66% for heart failure. Fig. 9f shows the cardiac mechanical and electrical biosignals captured by a hydrogel-based device during walking in a patient clinically diagnosed with heart failure. The signals show reduced Q and T peaks, irregular heart rate, interrupted electromechanical delay, and altered pre-ejection period/left ventricular ejection time parameters, indicating electrophysiological disturbances. Additionally, the patient's cardiac mechanical activity is significantly impaired, with a contraction amplitude nearly 60% lower than that of healthy individuals (Fig. 9g). These results demonstrate the ability of hydrogelbased epidermal electrodes to achieve high-fidelity recording of cardiac signals.

4.2 Electromyography monitoring

EMG is a technique for recording electrophysiological activity associated with muscle contraction and relaxation¹⁴⁵. As EMG signals directly reflect neuromuscular activity, they are extensively utilized in human motion monitoring¹⁴⁶⁻¹⁴⁸, controlling human-machine interfaces¹⁴⁹⁻¹⁵¹, and diagnosing neuromuscular disorders^{45, 152, 153}. However, the impedance between the electrode and skin affects the quality of EMG signals in practical applications, so researchers have increasingly focused on enhancing the conductivity and skin adhesion of hydrogel-based epidermal electrodes to achieve low interface impedance^{142, 154-157}. For instance, Tian et al.¹⁵⁸ leveraged the liquid-to-solid transition concept to develop an in-situ hydrogel-based epidermal electrode for monitoring post-fatigue muscle recovery (Fig. 10a). To develop a hydrogel with rapid in situ gelation, a formulation of gelatin, PEDOT:PSS, and a deep eutectic solvent (DES) was employed. At temperatures above 60 °C, gelatin exists as individual molecular

chains. Upon cooling, the gelatin network solidifies via hydrogen bonding and physical phosical crosslinking, with PEDOT:PSS forming a conductive network within the gelatin matrix (Fig. 10b). DES demonstrate superior thermal stability and ionic conductivity. The hydroxyl groups in DES engage with the polymer network via hydrogen bonding and ionic interactions, improving solubility, stability, and network formation. The in situ-formed hydrogel exhibits exceptional adhesion (591 kPa) and minimal skin contact impedance (10.2 k Ω). The prepared in-situ biogel epidermal electrode was used to

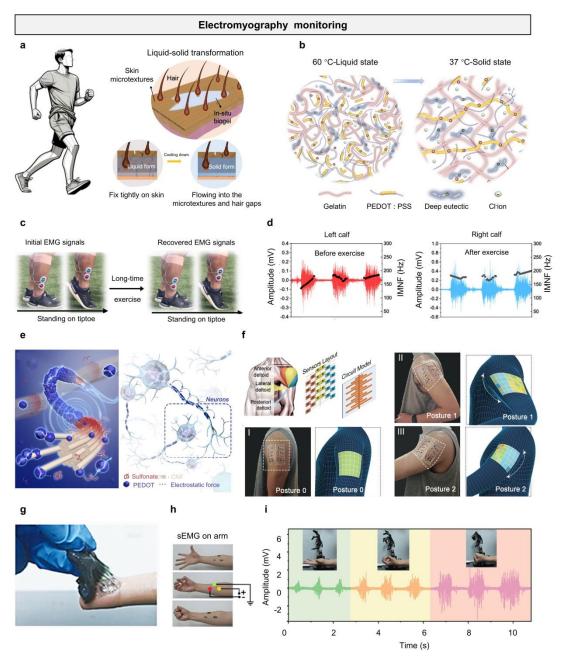


Fig. 10 Electromyography monitoring. (a) Liquid-to-solid transformation concept and the application of in situ biogel. (b) Schematic diagram and optical images of the

713

714

715

716

717

718

719

720

721

722

723

724

725

726

727

728

729

730

731

732

733

734

735

736

737

738

739

740

741

in situ rapid gelation process of the biogel. (c) Procedure for recovery monitoring using View Article Online Distriction of the biogel. in situ biogel. Comparison of initial EMG signals with those after fatigue. (d) Initial EMG signals and post-exercise signals for left and right calves. Reproduced with permission [158]. Copyright 2025, Nature Publishing Group. (e) Schematic illustration of the biomimetic segmentally conductive fiber hydrogel. (f) Sensor array layout and circuit model designed for monitoring muscle movements. (i) Photograph of the selfadhesive hydrogel after peeling. (h, i) EMG signals on the forearm at different grip force levels. Reproduced with permission [99]. Copyright 2024, American Chemical Society. monitor muscle recovery after exercise-induced fatigue. EMG signals from the left and right calves were recorded when volunteers performed toe-raising exercises (Fig. 10c). After prolonged exercise (running for 45 minutes), the hydrogel epidermal electrode recorded EMG signals at different stages, with an SNR of up to 30 dB (Fig. 10d). Hydrogel-based epidermal electrodes were employed to monitor muscle signals in volunteers during extended sedentary periods, revealing a decline in average muscle frequency over time. After 24 hours, the mean muscle frequency started to rise, approaching pre-exercise levels by 48 hours, signifying full recovery from exerciseinduced fatigue. To quantitatively assess muscle fatigue recovery, the recovery percentage was defined as the ratio of mean EMG frequencies before and after exerciseinduced fatigue. This recovery metric enables precise evaluation of muscle condition, supporting exercise and recovery planning, as well as monitoring recovery from muscle-related disorders. Liu et al.99 designed high-performance hydrogel-based epidermal electrodes by incorporating numerous interpenetrating core-sheath conductive nanofibers into a physically crosslinked polyelectrolyte network. The hydrogel is composed of cellulose nanofibers, PAA, and PEDOT:PSS (Fig. 10e). The hydrogel-based epidermal electrode was fabricated as a 10 × 10 cm² multi-pixel skin array, showcasing its capability to capture human motion across various postures (Fig. 10f). As an interface for human-machine interaction, the hydrogel epidermal electrode exhibits good interfacial adhesion (adhesion energy of 28 kPa on pig skin) and a low electrochemical impedance of 42 Ω . Fig. 10g shows that after peeling from the skin, a typical "fibrillation" phenomenon occurs, confirming effective adhesion between the

743

744

745

746

747

748

749

750

751

752

753

754

755

756

757

758

759

760

761

762

763

764

765

766

767

768

View Article Online

hydrogel epidermal electrode and the skin (Fig. 10h). To evaluate the human-machine/D5TC018963 interaction capability of the hydrogel epidermal electrode, the electrode was used to test EMG signals on the forearm under different grip strength levels, and the recorded arm EMG signals could be customized to precisely control a prosthetic limb, thereby achieving various robotic gestures with adjustable grasping angles and force levels (Fig. 10i).

4.3 Electroencephalogram monitoring

EEG is an electrophysiological monitoring method for recording brain electrical activity. 159 It mainly originates from postsynaptic potentials and has significant application potential in sleep monitoring 160-162, emotion recognition 163-165, and braincomputer interfaces 166-168. The presence of scalp hair and the head's intricate, curved contours pose significant challenges in achieving stable and effective contact between epidermal electrodes and the scalp for prolonged EEG recordings¹⁶⁹. To overcome this challenge, Wang et al. 170 developed a conductive gel-based epidermal electrode for skin application, featuring a temperature-responsive reversible liquid-to-gel transition. This phase transition endows the hydrogel with unique skin coating and in-situ gelation properties (Fig. 11a). The fluidity of the conductive hydrogel enables good compliance and conformal contact with hairy scalps without hair interference, surpassing prefabricated gels, which facilitates high-quality EEG recording through hairy scalps (Fig. 11b). The conductive hydrogel-based epidermal electrode was utilized to capture EEG signals during open-eye and closed-eye conditions, revealing distinct differences between the signals in these paradigms. Due to its relatively long-term electrical stability, excellent mechanical interaction (73.4 \pm 2.2 kPa), and stable contact with the scalp $(6.95 \pm 0.97 \text{ k}\Omega)$, the conductive hydrogel epidermal electrode recorded α signals with no significant differences across different wearing durations (0, 24, 48, and 72 hours), with high quality and a peak frequency of 10 Hz (Fig. 11c). This indicates that the applicable conductive hydrogel epidermal electrode has promising capabilities for

long-term high-fidelity EEG recording. To enable continuous high-fidelity EE@signat/D5TC01896J detection, paintable hydrogel-based epidermal electrodes require a streamlined and rapid gelation process to minimize adhesion-related damage from slow gelation and reduce subject discomfort. Li et al. ¹⁷¹developed a paintable, fast-gelling, and highly adhesive hydrogel-based epidermal electrode via a one-pot synthesis. Comprising gelatin, gallic acid, sodium citrate, LiCl, glycerol, and Tris-HCl buffer (Gel-GGLiCit),

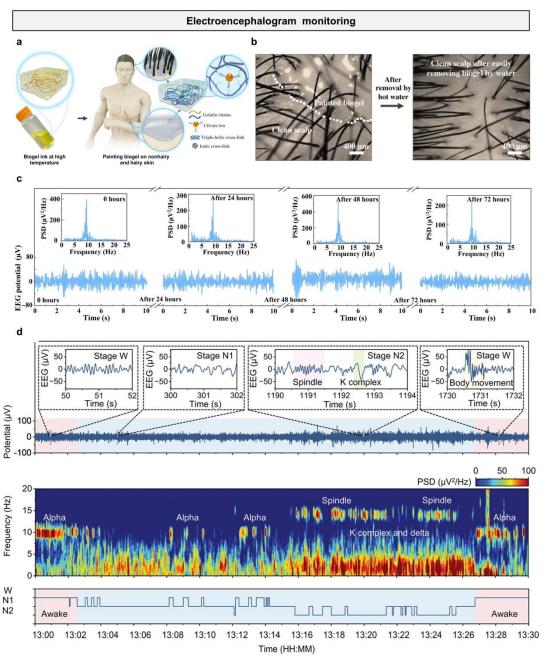


Fig. 11 Electroencephalogram monitoring. (a) Schematic illustration showing the concept of on-skin paintable biogel for hairy scalp for EEG recording. (b) Optical images of biogel painted on the hairy scalp and clean scalp after removing biogel by

797

798

799

800

801

802

803

804

805

806

807

This article is licensed under a Creative Commons Attribution-NonCommercial 3.0 Unported Licence

Open Access Article. Published on 05 2025. Downloaded on 29/10/2025 01:38:06.

water. (c) EEG alpha rhythms recorded by the painted biogel electrode after continuous/D5TC01896J wearing for 0, 24, 48, and 72 hours. Reproduced with permission [170]. Copyright 2022, American Association for the Advancement of Science. (d) Continuous wireless monitoring of EEG signals using paintable Gel-GGLiCit hydrogels and the multitaper spectrogram of the EEG signals (top) and visually scored hypnogram (bottom). Reproduced with permission [171]. Copyright 2024, Wiley-VCH. the hydrogel possesses reversible thermal phase transition properties, facilitating skin compatibility and rapid in situ gelation within 15 seconds. Furthermore, the paintable hydrogel-based epidermal electrode can form a conformal interface with hairy scalps (3.36 mJ/cm²) in its fluid state prior to gelation, allowing it to penetrate dense hair effectively. These properties substantially lower scalp contact impedance, with the electrode achieving a reduced skin contact impedance of 45.64 kΩ compared to 62.70 kΩ for commercial pastes. To assess the potential of the paintable hydrogel-based epidermal electrode for sleep stage monitoring and sleep disorder analysis, it was employed to capture EEG signals from subjects during brief daytime naps. In addition to the awake state, the subject's 30-minute sleep primarily involved non-rapid eye movement (NREM) sleep, stages N1 and N2. Stage N1 marks the initial phase of sleep, defined by the transition from wakefulness to sleep, where individuals experience light sleep and are easily roused. Following N1, stage N2 emerges as a deeper sleep phase, characterized by a reduced heart rate and the occurrence of sleep spindles (Fig. 11d). During the awake stage (W), elevated power in the alpha band (8–12 Hz) is evident in EEG signals, most pronounced during eye closure. As wakefulness transitions to stage N1, alpha band power progressively diminishes and eventually disappears. These findings demonstrate that high-fidelity raw EEG data, acquired with paintable hydrogel-based epidermal electrodes, enable clear visualization of critical EEG features throughout sleep.

4.4 Electrooculography monitoring

EOG is a technique that measures eye movement and position by recording electrical potentials around the eyes, widely used for diagnosing ophthalmic and neurological

View Article Online

808

809

810

811

812

813

814

815816

817

818 819 disorders^{172, 173}, supporting human-computer interaction^{174, 175}, studying cognitive/DSTC018963 processes^{176, 177}, and assessing fatigue and alertness^{178, 179}. Zheng et al.¹⁸⁰ developed a thermoresponsive gelatin-based hydrogel (GGW) via a one-pot synthesis, incorporating glycerol, ammonium chloride (NH₄Cl), and water (Fig. 12a). The in situ-formed hydrogel electrode demonstrates robust adhesion at the hydrogel-skin interface (0.9 N/cm), enabling conformal skin contact and high-fidelity signal acquisition (Fig. 12b).

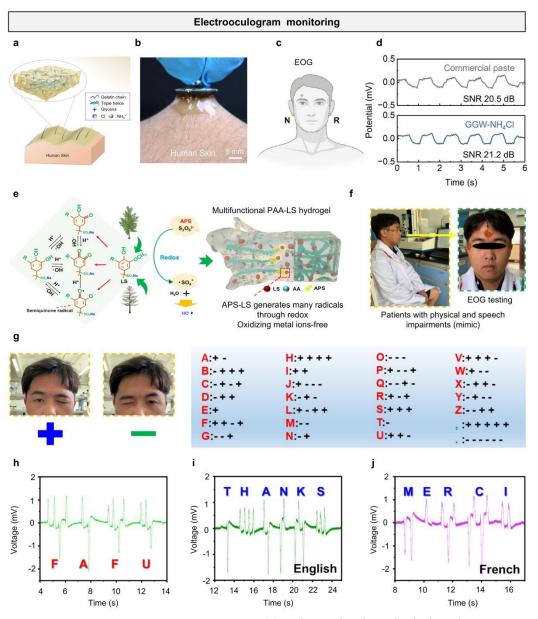


Fig. 12 Electrooculography monitoring. (a) Schematic view depicting the concept of a skin-printable hydrogel. (b) Photograph showing the attachment of Ag/AgCl electrode to the GGW-NH₄Cl hydrogel, highlighting the superior adhesion. (c) Schematic of electrode positions for EOG signal measurements. (d) EOG signals recorded using commercial paste (top) and the GGW-NH4Cl hydrogel (bottom), along

with the corresponding SNR. Reproduced with permission [180]. Copyright 2025 plost Colleged SNR. Reproduced with permission [180]. Copyright 2025 plost Colleged SNR. Reproduced with permission [180]. Copyright 2025 plost Colleged SNR. Reproduced with permission [180]. Copyright 2025 plost Colleged SNR. Reproduced with permission [180]. Copyright 2025 plost Colleged SNR. Reproduced with permission [180]. Copyright 2025 plost Colleged SNR. Reproduced with permission [180]. Copyright 2025 plost Colleged SNR. Reproduced with permission [180]. Copyright 2025 plost Colleged SNR. Reproduced with permission [180]. Copyright 2025 plost Colleged SNR. Reproduced hydrogels. (f) Schematic diagram of PAA-LS-0.05 hydrogel-based bioelectrodes adhered to the skin of a volunteer's head for EOG monitoring. (g) Schematic diagram of the codable eye communication mechanism. (h) The gel electrodes detected the volunteers' eye movements, which the device interpreted as the word "FAFU". This volunteer effectively produced the outputs i)

"thanks" and j) "merci" (French). Reproduced with permission [181]. Copyright 2025,

828 829

827

830

831

832

833

834

835

836

837

838

839

840

841

842

843

844

845

846

847

848

849

850

851

Elsevier. The hydrogel was placed around the eyes for EOG signal monitoring, and then the signals were transmitted through a Bluetooth-enabled portable wireless module for realtime data collection. Fig. 12c illustrates representative EOG signals acquired with hydrogel-based epidermal electrodes, exhibiting pronounced periodic waveforms as the subject repeatedly shifts their gaze from side to side. Notably, the EOG signals obtained using the hydrogel epidermal electrode exhibit higher amplitude than those recorded using commercial paste. Meanwhile, the SNR (21.2 dB) of the hydrogel epidermal electrode surpasses that of the commercial paste (20.5 dB) (Fig. 12d). To validate the capability of the hydrogel epidermal electrode in acquiring EOG signals from patients with speech impairments, Pan et al. reported a multifunctional PAA hydrogel catalyzed by oxidative metal-ion-free lignin¹⁸¹. By incorporating transition metal ions and sodium lignosulfonate (LS), LS rapidly reduces Fe³⁺ to Fe²⁺, thereby promoting the rapid generation of free radicals by APS (initiator) at room temperature. This mechanism accelerates the polymerization of vinyl monomers, enabling the rapid assembly of the hydrogel (Fig. 12e). The hydrogel exhibits excellent flexibility (~7 kPa), remarkable stretchability (~2700% strain), and strong skin adhesion (~9 kPa). Notably, the hydrogel, integrating these superior properties, can serve as an epidermal electrode for the human body, offering a viable alternative to conventional commercial electrodes. Fig. 12f illustrates a volunteer wearing the PAA-LS-0.05 hydrogel epidermal electrode undergoing an EOG test. The volunteer simulated a patient with severe physical and verbal impairments, retaining only limited facial skin mobility and lacking the ability to speak or type. To address this, the actions of closing the left and

853

854

855

856

857

858

859

860

861

862

863

864

865

866

867

868

869

870

871

872

873

874

875

876

877

right eyes were defined as "+" and "-", respectively (Fig. 12g), and a reference table/D5TC01896J comprising 26 English letters and punctuation marks was developed. Through regular eye movements, the patient could achieve linguistic output and communication. As shown in Fig. 12h, the patient successfully produced the unordered sequence "F A F U" via eye movements, with the signal being clearly readable. Subsequently, the patient further expressed the English word "THANKS" (Fig. 12i) and the French word "MERCI" (meaning "thank you", Fig. 12j) through eye movements. This wearable hydrogel electrode seamlessly integrates with the eye-movement-based communication system, offering a significant and urgent solution to address communication challenges for such patients. While conductive hydrogel-based epidermal electrodes have demonstrated excellent performance across various electrophysiological modalities—including ECG, EMG, EEG, and EOG—their practical deployment in dynamic, real-world environments poses additional challenges related to signal stability and quality. Motion artifacts remain a major concern in wearable and long-term bioelectronic monitoring, often arising from skin deformation, electrode displacement, or body movement. These artifacts can significantly distort low-amplitude biopotentials—particularly in EEG and ECG recordings—thereby compromising signal fidelity and diagnostic accuracy. The intrinsic softness, stretchability, and conformability of hydrogel-based electrodes help maintain stable skin-electrode contact, effectively minimizing impedance fluctuations and suppressing artifact generation at the source. Additionally, signal processing techniques such as adaptive filtering, baseline drift correction, and motion-sensorassisted denoising are widely employed to further improve signal quality. Therefore, the integration of material-level optimization and algorithmic post-processing is essential for achieving reliable, high-fidelity electrophysiological recordings under real-life conditions.

This article is licensed under a Creative Commons Attribution-NonCommercial 3.0 Unported Licence.

Open Access Article. Published on 05 2025. Downloaded on 29/10/2025 01:38:06.

View Article Online DOI: 10.1039/D5TC01896J

5. Summary and Perspectives

Conductive hydrogels, characterized by their intrinsic flexibility, tunable properties on adhesion and electrical conductivity, hold significant promise for the development of high-performance epidermal electrodes. This review offers a comprehensive overview of recent advances in conductive hydrogel-based electrodes for electrophysiological monitoring. We first examined the underlying conductive mechanisms—ionic, electronic, and hybrid pathways—followed by a discussion of the essential properties required for effective signal acquisition, such as high conductivity, strong adhesion, stretchability, and breathability. Finally, we highlighted recent progress in their application across various electrophysiological monitoring scenarios. Despite notable achievements, several challenges persist that must be addressed to fully realize their potential.

Advancements in materials and conductivity mechanisms

The future development of conductive hydrogel-based epidermal electrodes will be driven by innovations in materials science and the continued refinement of conductivity mechanisms. While current strategies largely focus on ionic, electronic, and hybrid conduction, future research will likely prioritize optimizing these mechanisms for enhanced signal fidelity and long-term stability. Incorporating advanced conductive polymers and nanomaterials could significantly improve electronic conductivity, overcoming the environmental sensitivity associated with ionic pathways. Additionally, bioinspired designs—such as the integration of conductive peptides or materials that mimic the skin's natural electrical properties—offer the potential for improved biocompatibility and seamless integration with biological systems. These innovations aim to produce electrodes capable of reliable, high-quality electrophysiological monitoring in dynamic and diverse conditions.

Strategies for long-term reliability

Long-term performance and durability of conductive hydrogel-based epidermal

electrodes can be greatly enhanced through advanced materials design and interfacial posterior process of the development of fatigue-resistant architectures, such as self-healing polymer networks and dynamic crosslinking systems, which can maintain mechanical integrity under repeated deformation. Hybrid structures that combine elastomers with hydrogels may further improve stretchability and resistance to cracking. To address adhesion challenges under real-world conditions—such as perspiration, oily skin, or continuous body motion—bioinspired adhesive strategies present an exciting pathway. These may include catechol-functionalized chemistries, microstructured surface designs, or reversible covalent bonding, enabling robust and repeatable skin contact. Advances in hydration management are also expected to play a crucial role in maintaining performance over extended use. Future studies may explore the integration of moisture-retentive additives, breathable yet protective encapsulation materials, or systems capable of autonomous hydration regulation to prevent dehydration and ion leaching.

Multifunctionality and smart features

Beyond signal monitoring, the next generation of conductive hydrogel electrodes is expected to embrace multifunctionality and smart responsiveness. One promising direction is the development of closed-loop systems, where electrodes not only detect physiological signals but also trigger therapeutic interventions, such as on-demand drug delivery. Incorporating self-healing capabilities through dynamic covalent bonds or supramolecular interactions could greatly enhance durability, especially under mechanical strain during long-term use. Additional smart features—such as energy harvesting from body movement or adaptive behavior in response to skin conditions—could further extend the functionality and lifespan of these devices. These advancements will transform conductive hydrogels into versatile, intelligent platforms that bridge sensing, therapy, and human-machine interfaces.

Wearability, data integration, and personalized medicine

The long-term vision for conductive hydrogel-based epidermal electrodes lies in their

Open Access Article. Published on 05 2025. Downloaded on 29/10/2025 01:38:06.

This article is licensed under a Creative Commons Attribution-NonCommercial 3.0 Unported Licence.

seamless integration into wearable systems that support comfort, continuous data //D5TC018963 acquisition, and personalized healthcare. Innovations in material design—particularly ultrathin, breathable, and skin-conformal hydrogels—will be crucial to improving wearability and minimizing skin irritation during extended use. Simultaneously, integrating these devices with advanced data analytics and machine learning algorithms will unlock their potential for real-time monitoring, predictive diagnostics, and personalized treatment strategies. This convergence of soft materials, digital health, and precision medicine heralds a shift toward proactive, patient-centered care, where electrophysiological monitoring becomes not just reactive but a key driver in managing health and disease.

CRediT Authorship Contribution Statement

Jiawei Yang: Writing – origin draft, Writing – review&editing, Investigation, Data curation. Yi Liu: Writing – origin draft, Writing – review&editing, Resources, Methodology, Data curation. Wenqing Yan: Writing – review&editing, Formal analysis. Pengcheng Zhou: Writing – review&editing, Formal analysis. Zonglei Wang: Writing – review&editing, Formal analysis. Yuli Wang: Writing – review&editing. Yujie Zhang: Writing – review&editing, Formal analysis. Zongman Zhang: Writing – review&editing, Software. Fan Mo: Writing – review&editing. Zichong Ji: Writing – review&editing. Hossam Haick: Writing – review&editing. Yan Wang: Writing – review&editing, Project administration, Funding acquisition, Conceptualization.

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.

View Article Online DOI: 10.1039/D5TC01896J

Acknowledgments

- The authors sincerely acknowledge the support from the Natural Science Foundation
- 959 of China (grant No.: 52303371, W2521021), Guangdong Science and Technology
- 960 Department (grant Nos.: STKJ2023075, 2022A1515110209, 2021B0301030005), seed
- 961 fund from GTIIT Changzhou Innovation Institute (grant No.: GCII-Seed-202406), and
- the Key Discipline (KD) Fund, the Technion, and the Start-Up Fund from Guangdong
- 963 Technion.

964

957

Notes and Reference

- Y. Xiang, K. Shi, Y. Li, J. Xue, Z. Tong, H. Li, Z. Li, C. Teng, J. Fang and N.
 Hu, *Nano-Micro Lett.*, 2024, 16, 132.
- J. P. Piccini, A. M. Russo, P. S. Sharma, J. Kron, W. Tzou, W. Sauer, D. S.
 Park, U. Birgersdotter-Green, D. S. Frankel and J. S. Healey, *Circulation:* Arrhythmia and Electrophysiology, 2022, 15, e009911.
- K. E. Odening, A.-M. Gomez, D. Dobrev, L. Fabritz, F. R. Heinzel, M. E.
 Mangoni, C. E. Molina, L. Sacconi, G. Smith and M. Stengl, *EP Europace*,
 2021, 23, 1795-1814.
- A. H. Caillet, A. T. Phillips, L. Modenese and D. Farina, *J. Electromyogr. Kinesiol.*, 2024, **76**, 102873.
- 975 5. R. Sun, A. Sohrabpour, G. A. Worrell and B. He, *PNAS*, 2022, **119**, 976 e2201128119.
- 977 6. F. Arcuri, C. Porcaro, I. Ciancarelli, P. Tonin and A. Cerasa, *Electronics*, 2021, 978 **10**, 836.
- 979 7. M. Pyasik, M. Scandola and V. Moro, Neuropsychologia, 2022, 174, 108333.
- 980 8. C.-l. a. R. Labs, D. Sussillo, P. Kaifosh and T. Reardon, *Biorxiv*, 2024, 981 2024.2002. 2023.581779.
- H. Zhang, D. Zhang, Z. Wang, G. Xi, R. Mao, Y. Ma, D. Wang, M. Tang, Z.
 Xu and H. Luan, ACS Appl. Mater. Interfaces, 2023, 15, 5128-5138.
- 984 10. J. W. Kam, T. Rahnuma, Y. Park and C. Hart, *Neuroimage*, 2022, **258**, 119372.
- 985 11. M. Zhu, H. Wang, S. Li, X. Liang, M. Zhang, X. Dai and Y. Zhang, *Adv. Healthcare Mater.*, 2021, **10**, 2100646.
- 987 12. Y. Luo, W. Li, Q. Lin, F. Zhang, K. He, D. Yang, X. J. Loh and X. Chen, *Adv. Mater.*, 2021, **33**, 2007848.
- 989 13. P. Kateb, J. Fan, J. Kim, X. Zhou, G. A. Lodygensky and F. Cicoira, *Flexible Printed Electron.*, 2023, **8**, 045006.

- J. Kim, J. Fan, G. Petrossian, X. Zhou, P. Kateb, N. Gagnon-Lafrenais and Figure Online Onlin
- 393 15. X. Zhou, P. Kateb, J. Fan, J. Kim, G. A. Lodygensky, B. Amilhon, D. Pasini
 394 and F. Cicoira, *J. Mater. Chem. C*, 2024, 12, 5708-5717.
- 995 16. F. Han, X. Huang and E. Teye, *J. Food Process Eng*, 2019, **42**, e12983.
- 996 17. Q. Ouyang, Y. Yang, J. Wu, Q. Chen, Z. Guo and H. Li, Lwt, 2020, 118, 108768.
- 997 18. W. Zhang, C. Liu, F. Liu, X. Zou, Y. Xu and X. Xu, *Food Chem.*, 2020, **303**, 998 125378.
- 999 19. B. Lu, F. Han, J. H. Aheto, M. M. Rashed and Z. Pan, *Food Science & Nutrition*, 2021, **9**, 5220-5228.
- 1001 20. H. Yin, X. Hu, X. Huang, X. Zou, Y. Xu, J. Shi and M. Yang, Food Anal.
 1002 Methods, 2021, 14, 1836-1842.
- 1003 21. S. Yang and X. Jiang, ACS Nano, 2024, 18, 27107-27125.
- 1004 22. H. Ullah, M. A. Wahab, G. Will, M. R. Karim, T. Pan, M. Gao, D. Lai, Y. Lin and M. H. Miraz, *Biosensors*, 2022, **12**, 630.
- J. Yi, Y. Gu, J. Yang, Z. Wang, Y. Wang, W. Yan, Q. Sun, P. Zhou, Y. Xu, X.
 He, J. Zhong and Y. Wang, *Mater. Horiz.*, 2025, DOI: 10.1039/D4MH01858C.
- J. Yang, Q. Sun, Z. Wang, Y. Xu, Y. Wang, W. Yan, P. Zhou, Z. Ji, H. Jiang,
 S. Chen, W. Zhang, H. Haick and Y. Wang, Wearable Electronics, 2025, 2, 55-
- 1010 61.
- 1011 25. H. Wu, G. Yang, K. Zhu, S. Liu, W. Guo, Z. Jiang and Z. Li, *Adv. Sci.*, 2021, **8**, 1012 2001938.
- 1013 26. L. Hu, P. L. Chee, S. Sugiarto, Y. Yu, C. Shi, R. Yan, Z. Yao, X. Shi, J. Zhi and D. Kai, *Adv. Mater.*, 2023, **35**, 2205326.
- 1015 27. L. Wang, T. Xu and X. Zhang, TrAC Trends Anal. Chem., 2021, 134, 116130.
- 1016 28. M. L. Oyen, *Int. Mater. Rev.*, 2014, **59**, 44-59.
- 1017 29. E. M. Ahmed, J. Adv. Res., 2015, 6, 105-121.
- 1018 30. H. Yuk, B. Lu and X. Zhao, Chem. Soc. Rev., 2019, 48, 1642-1667.
- 1019 31. Y. Zhao, X. Fu, B. Liu, J. Sun, Z. Zhuang, P. Yang, J. Zhong and K. Liu, *Sci. China. Mater.*, 2023, **66**, 1934-1940.
- 1021 32. F. Mo, P. Zhou, S. Lin, J. Zhong and Y. Wang, *Adv. Healthcare Mater.*, 2024, 1022 13, 2401503.
- 1023 33. Y. Huang, M. Xiao, X. Zhou, J. Zhu, Y. Tian, S. Xie, Y. Gong and J. Zhong, 1024 Sens. Actuators B Chem., 2025, **431**, 137461.
- 1025 34. Y. Liu, L. Han, S. Lv, T. Jiang, M. Duan, H. Guo, Y. Li, Q. Xie, Y. Chen and
 1026 D. Wang, *Research*, 2025, 8, 0714.
- 1027 35. Y. Zhou, Y. Zhao, D. Zhao, X. Guan, K. Zhang, Y. Pi and J. Zhong, *Microsyst*. 1028 *Nanoeng.*, 2025, **11**, 40.
- 1029 36. K. Deligkaris, T. S. Tadele, W. Olthuis and A. van den Berg, *Sens. Actuators B Chem.*, 2010, **147**, 765-774.
- 1031 37. Y. S. Zhang and A. Khademhosseini, *Science*, 2017, **356**, eaaf3627.
- 1032 38. P. Zhou, F. Mo, Z. Ji, J. Yang, H. Du, Z. Wang, H. Haick and Y. Wang, Sci.

- Open Access Article. Published on 05 2025. Downloaded on 29/10/2025 01:38:06.

 This article is licensed under a Creative Commons Attribution-NonCommercial 3.0 Unported Licence.
- 1033 Bull., 2025, DOI: https://doi.org/10.1016/j.scib.2025.01.058.
- View Article Online DOI: 10.1039/D5TC01896J
- 1034 39. F. Mo, P. Zhou, S. Lin, J. Zhong and Y. Wang, *Adv. Healthcare Mater.*, 2024, 1025
- 1035 **13**, 2401503.
- 1036 40. P. Zhou, Z. Zhang, F. Mo and Y. Wang, Adv. Sens. Res., 2024, 3, 2300021.
- 1037 41. F. Mo, Y. Liu, P. Zhou, J. Yang, Z. Ji and Y. Wang, *Mater. Sci. Eng. R* 1038 *Rep.*, 2025, **165**, 100989.
- 1039 42. M. Niu, K. Chen, W. Li, J. Hu, J. Zhang, P. Zhu, Z. Pan and Y. Mao, *J. Mater.* 1040 *Res.*, 2024, **39**, 188-211.
- 1041 43. H. Duan, Y. Zhang, Y. Zhang, P. Zhu and Y. Mao, *Nanomaterials*, 2024, **14**, 1042 1398.
- 1043 44. W. Hu, D. Song, X. Shi and N. Liu, *SCIENTIA SINICA Chimica*, 2022, **52**, 837-1044 847.
- 1045 45. H. Ding, Y. Gu, Y. Ren, C. Hu, Q. Qiu, D. Wu, J. Mou, Z. Wu and H. Zhou, *J. Mater. Chem. C*, 2024.
- 1047 46. Q. Han, C. Zhang, T. Guo, Y. Tian, W. Song, J. Lei, Q. Li, A. Wang, M. Zhang,
 1048 S. Bai and X. Yan, *Adv. Mater.*, 2023, 35, 2209606.
- 1049 47. Q. Han, X. Gao, C. Zhang, Y. Tian, S. Liang, X. Li, Y. Jing, M. Zhang, A. Wang and S. Bai, *Adv. Mater.*, 2025, **37**, 2415445.
- 48. Y. Li, Y. Gu, S. Qian, S. Zheng, Y. Pang, L. Wang, B. Liu, S. Liu and Q. Zhao,
 Nano Res., 2024, 17, 5479-5490.
- 1053 49. Z. Zhang, J. Yang, H. Wang, C. Wang, Y. Gu, Y. Xu, S. Lee, T. Yokota, H.
 1054 Haick, T. Someya and Y. Wang, *Sci. Adv.*, 2024, 10, eadj5389.
- T.-C. Ho, C.-C. Chang, H.-P. Chan, T.-W. Chung, C.-W. Shu, K.-P. Chuang,
 T.-H. Duh, M.-H. Yang and Y.-C. Tyan, *Molecules*, 2022, 27, 2902.
- T. Zhu, Y. Ni, G. M. Biesold, Y. Cheng, M. Ge, H. Li, J. Huang, Z. Lin and Y.
 Lai, Chem. Soc. Rev., 2023, 52, 473-509.
- 1059
 52. H. Dechiraju, M. Jia, L. Luo and M. Rolandi, Adv. Sustain. Syst., 2022, 6,
 1060
 2100173.
- S. Chen, Y. Chen, X. Mu, P. Wang, L. Miao, S. Tanemura and H. Cai, *Sustain*.
 Mater. Technol., 2023, 36, e00635.
- X. Li, Y. Sun, S. Wang, G. Tian, T. Yang, L. Huang, Y. Ao, B. Lan, J. Zhang,
 T. Xu, Y. Liu, L. Jin, W. Yang and W. Deng, *Chem. Eng. J.*, 2024, 498, 155195.
- 1065 55. C. G. Wang, N. E. B. Surat'man, J. J. Chang, Z. L. Ong, B. Li, X. Fan, X. J. Loh and Z. Li, *Chemistry—An Asian Journal*, 2022, **17**, e202200604.
- 1067 56. A. S. Ivanov, L. V. Pershina, K. G. Nikolaev and E. V. Skorb, *Macromol.* 1068 *Biosci.*, 2021, **21**, 2100117.
- 1069 57. D. Lu, Z. Zhu, M. Zhu, P. Zhang and X. Xiang, *J. Mater. Chem. A*, 2025, **13**, 1070 427-440.
- 1071 58. Y. Gao, W. Zhang, L. Li, Z. Wang, Y. Shu and J. Wang, *Chem. Eng. J.*, 2023, 1072 **452**, 139248.
- 1073 59. Z. Luo, W. Li, J. Yan and J. Sun, Adv. Funct. Mater., 2022, 32, 2203988.
- 1074 60. Y. Zhao, F. Wang, J. Liu, D. Gan, B. Lei, J. Shao, W. Wang, Q. Wang and X.

View Article Online

- 1075 Dong, ACS Appl. Mater. Interfaces, 2023, 15, 28664-28674. DOI: 10.1039/D5TC01896J
- 1076 Q. He, Y. Cheng, Y. Deng, F. Wen, Y. Lai and H. Li, Adv. Funct. Mater., 2024, 61. 1077 **34**, 2308974.
- F. Miguel, F. Barbosa, F. C. Ferreira and J. C. Silva, Gels, 2022, 8, 710. 1078 62.
- 1079 M. A. Bhat, R. A. Rather and A. H. Shalla, Synth. Met., 2021, 273, 116709. 63.
- 1080 B. Zhao, Z. Li, L. Zheng, Z. Ye, Y. Yuan, S. Zhang, B. Liang and T. Li, Chin. 64. 1081 Chem. Lett., 2024, 35, 109810.
- 1082 R. Eivazzadeh-Keihan, E. B. Noruzi, E. Chidar, M. Jafari, F. Davoodi, A. 65.
- 1083 Kashtiaray, M. G. Gorab, S. M. Hashemi, S. Javanshir and R. A. Cohan, *Chem.*
- 1084 Eng. J., 2022, 442, 136183.
- 1085 66. R. Arambula-Maldonado and K. Mequanint, Materials Advances, 2022, 3, 1086 5186-5206.
- 1087 Y. Zhao, K. Zhao, R. Qian, Z. Yu and C. Ye, Chem. Eng. J., 2024, 150197. 67.
- 1088 68. Y. Wang, M. Zhang, Z. Yan, S. Ji, S. Xiao and J. Gao, *Theranostics*, 2024, 14, 1089 1534.
- 1090 69. X. Huang, C. Chen, X. Ma, T. Zhu, W. Ma, Q. Jin, R. Du, Y. Cai, M. Zhang, D.
- 1091 Kong, M. Wang, J. a. Ren, Q. Zhang and X. Jia, Adv. Funct. Mater., 2023, 33, 1092 2302846.
- 1093 70. J. Dai, D. Ren, S. Zhang, Y. Liu, Y. Xiao, Z. Wang, B. Wang and F. Huang, 1094 ACS Appl. Electron. Mater., 2025, 7, 3125-3134.
- 1095 Y. Ohm, C. Pan, M. J. Ford, X. Huang, J. Liao and C. Majidi, Nat. Electron., 71. 1096 2021, 4, 185-192.
- 1097 72. J. Chen, F. Liu, T. Abdiryim and X. Liu, Adv. Compos. Hybrid Mater., 2024, 7, 1098 35.
- 1099 73. J. Luo, C. Sun, B. Chang, Y. Jing, K. Li, Y. Li, Q. Zhang, H. Wang and C. Hou, 1100 ACS Nano, 2022, 16, 19373-19384.
- 1101 74. C. Yu, Z. Yue, H. Zhang, M. Shi, M. Yao, Q. Yu, M. Liu, B. Guo, H. Zhang, L. 1102 Tian, H. Sun, F. Yao and J. Li, Adv. Funct. Mater., 2023, 33, 2211023.
- P. Wang, Y. Lv, J. Duan, G. Sun, C. Meng, Y. Li, S. Guo and T. Zhang, Nano 1103 75. 1104 Energy, 2025, 136, 110722.
- 1105 76. F. Wang, L. Yang, Y. Sun, Y. Cai, X. Xu, Z. Liu, Q. Liu, H. Zhao, C. Ma and 1106 J. Liu, Gels, 2023, 9, 323.
- 1107 Z. Wang, L. Chen, Y. Chen, P. Liu, H. Duan and P. Cheng, Research, 2020, 77. 1108 **2020**.
- Y. Liu, C. Wang, J. Xue, G. Huang, S. Zheng, K. Zhao, J. Huang, Y. Wang, Y. 1109 78. Zhang and T. Yin, Adv. Healthcare Mater., 2022, 11, 2270092. 1110
- L. Rong, X. Xie, W. Yuan and Y. Fu, ACS Appl. Mater. Interfaces, 2022, 14, 1111 79. 1112 29273-29283.
- 1113 80. Y. Shi, Y. Ding, W. Wang and D. Yu, Colloids and Surfaces A: 1114 Physicochemical and Engineering Aspects, 2023, 675, 132081.
- R. Liu, T. Wang, G. Li, Z. Fan, Q. Zhou, K. Wang, P. Li and W. Huang, Adv. 1115 81. 1116 Funct. Mater., 2023, 33, 2214917.

- Q. Liang, X. Xia, X. Sun, D. Yu, X. Huang, G. Han, S. M. Mugo, W. Chen and Chotale Online Onl 82. 1117 1118
- Q. Zhang, Adv. Sci., 2022, 9, 2201059.
- 1119 Q. Wu, A. Chen, Y. Xu, S. Han, J. Zhang, Y. Chen, J. Hang, X. Yang and L. 83. Guan, Soft Matter, 2024, 20, 3666-3675. 1120
- X. Pan, Q. Wang, P. He, K. Liu, Y. Ni, X. Ouyang, L. Chen, L. Huang, H. Wang 1121 84. 1122 and Y. Tan, ACS Sustain. Chem. Eng., 2019, 7, 7918-7925.
- 1123 M. Lu, L. Shen, H. Su, B. Li, L. Wang and W. W. Yu, J. Colloid Interface Sci., 85. 1124 2025, **684**, 272-282.
- 1125 86. H. Ma, J. Hou, X. Xiao, R. Wan, G. Ge, W. Zheng, C. Chen, J. Cao, J. Wang,
- 1126 C. Liu, Q. Zhao, Z. Zhang, P. Jiang, S. Chen, W. Xiong, J. Xu and B. Lu, J.
- 1127 Colloid Interface Sci., 2024, **654**, 639-648.
- 1128 87. J. Lao, Y. Jiao, Y. Zhang, H. Xu, Y. Wang, Y. Ma, X. Feng and J. Yu, ACS 1129 *Nano*, 2025, **19**, 7755-7766.
- 1130 88. J. Zheng, J. Zhou, Y. Zhao, C. Wang, M. Fan, Y. Li, C. Yang and H. Yang, 1131 Biosensors, 2025, 15, 177.
- X. Xia, Q. Liang, X. Sun, D. Yu, X. Huang, S. M. Mugo, W. Chen, D. Wang 1132 89. and Q. Zhang, Adv. Funct. Mater., 2022, 32, 2208024. 1133
- 1134 90. H. Xue, D. Wang, M. Jin, H. Gao, X. Wang, L. Xia, D. a. Li, K. Sun, H. Wang, 1135 X. Dong, C. Zhang, F. Cong and J. Lin, *Microsyst. Nanoeng.*, 2023, 9, 79.
- 1136 91. N. Li, X. Wang, Y. Liu, Y. Li, J. Li, Z. Qin and T. Jiao, Chem. Eng. J., 2024, 1137 **483**, 149303.
- 1138 J. Wei, H. Chen, F. Pan, H. Zhang, K. Yang, T. Yuan, Y. Fang, H. Ping, Q. 92. 1139 Wang and Z. Fu, ACS Nano, 2025, 19, 15554-15564.
- 1140 93. X. Zhou, A. Rajeev, A. Subramanian, Y. Li, N. Rossetti, G. Natale, G. A. Lodygensky and F. Cicoira, Acta Biomater., 2022, 139, 296-306. 1141
- 1142 94. H. Huang, J. Shen, S. Wan, L. Han, G. Dou and L. Sun, ACS Appl. Mater. Interfaces, 2023, 15, 11549-11562. 1143
- 1144 95. D. Kim, H. J. Lee, J. Oh, H. Y. Yang, H. J. Park, C. Huh, D. H. Ha, Y. Jun and Y. J. Yun, J. Mater. Chem. C, 2025, 13, 5711-5718. 1145
- 1146 96. M. Li, W. Li, Q. Guan, J. Lv, Z. Wang, L. Ding, C. Li, E. Saiz and X. Hou, 1147 Device, 2023, 1.
- 1148 H. Su, L. Mao, X. Chen, P. Liu, J. Pu, Z. Mao, T. Fujiwara, Y. Ma, X. Mao and 97. 1149 T. Li, Adv. Sci., 2024, 11, 2405273.
- 1150 B. Yao, Y. Yan, Q. Cui, S. Duan, C. Wang, Y. Du, Y. Zhao, D. Wu, S. Wu, X. 98. 1151 Zhu, T. Hsiai and X. He, *Matter*, 2022, **5**, 4407-4424.
- C. Liu, Y. Wang, S. Shi, Y. Zheng, Z. Ye, J. Liao, Q. Sun, B. Dang and X. Shen, 1152 99. 1153 ACS Nano, 2024, 18, 27420-27432.
- 1154 100. H. He, H. Li, A. Pu, W. Li, K. Ban and L. Xu, Nat. Commun., 2023, 14, 759.
- 1155 101. J. Zhang, Y. Wang, Q. Wei, Y. Wang, M. Lei, M. Li, D. Li, L. Zhang and Y. 1156 Wu, Gels, 2021, 7, 216.
- Z. Chen, Y. Chen, M. S. Hedenqvist, C. Chen, C. Cai, H. Li, H. Liu and J. Fu, 1157 102. 1158 J. Mater. Chem. B, 2021, 9, 2561-2583.

- 1159 103. W. Li, J. Liu, J. Wei, Z. Yang, C. Ren and B. Li, *Adv. Funct. Mater.*, 2023, 33 p/D5TC018965 1160 2213485.
- 1161 104. J. Zhang, Y. Hu, L. Zhang, J. Zhou and A. Lu, *Nano-Micro Lett.*, 2022, **15**, 8.
- 1162 105. G. Kougkolos, M. Golzio, L. Laudebat, Z. Valdez-Nava and E. Flahaut, *J. Mater. Chem. B*, 2023, **11**, 2036-2062.
- 1164 106. L. Li, J. Meng, M. Zhang, T. Liu and C. Zhang, *Chem. Commun. (Cambridge, U. K.)*, 2022, **58**, 185-207.
- 1166 107. Z. Wang, X. Xu, R. Tan, S. Zhang, K. Zhang and J. Hu, *Adv. Funct. Mater.*, 1167 2024, **34**, 2312667.
- 1168 108. Q. Zhang, H. Lu, G. Yun, L. Gong, Z. Chen, S. Jin, H. Du, Z. Jiang and W. Li, 1169 *Adv. Funct. Mater.*, 2024, **34**, 2308113.
- 1170 109. S. Li, Y. Cong and J. Fu, J. Mater. Chem. B, 2021, 9, 4423-4443.
- 1171 110. X. Ma, X. Zhou, J. Ding, B. Huang, P. Wang, Y. Zhao, Q. Mu, S. Zhang, C.
 1172 Ren and W. Xu, J. Mater. Chem. A, 2022, 10, 11823-11853.
- 1173 111. X. Shi and P. Wu, Small, 2021, 17, 2101220.
- 1174 112. X. Shi and P. Wu, Small, 2021, 17, 2101220.
- 1175 113. Y. Gao, K. Wu and Z. Suo, Adv. Mater., 2019, 31, 1806948.
- 1176 114. T. Wang, P. Zhang, X. Yang, Y. Zhang, J. Zhang, X. He, P. Gu, X. Gong and Y. Zhao, *Chem. Eng. J.*, 2022, **438**, 135441.
- 1178 115. Z. Xu, X. Liang, W. Ma, X. An, H. Wu, Q. Zhang and X. Jia, *Adv. Funct. Mater.*, 1179 2024, **34**, 2310233.
- 1180 116. Y. Liu, P. Wang, X. Su, L. Xu, Z. Tian, H. Wang, G. Ji and J. Huang, *Adv. Mater.*, 2022, **34**, 2108820.
- 1182 117. G. Bovone, O. Y. Dudaryeva, B. Marco-Dufort and M. W. Tibbitt, *ACS Biomater. Sci. Eng.*, 2021, 7, 4048-4076.
- 1184 118. Y. Zhao, S. Song, X. Ren, J. Zhang, Q. Lin and Y. Zhao, *Chem. Rev.*, 2022, 1185 **122**, 5604-5640.
- 1186 119. Y.-W. Lee, S. Chun, D. Son, X. Hu, M. Schneider and M. Sitti, *Adv. Mater.*, 2022, **34**, 2109325.
- 1188 120. L. Nicolle, C. M. Journot and S. Gerber-Lemaire, *Polymers*, 2021, **13**, 4118.
- 1189 121. W. Zhang, Y. Zhang, Y. Dai, F. Xia and X. Zhang, J. Mater. Chem.
 1190 B, 2021, 9, 5954-5966.
- 1191 122. X. Wei, Y. Wang, Y. Liu, K. Ji, K. Li, J. Wang and Z. Gu, *Matter*, 2024, **7**, 826-1192 854.
- 1193 123. S. Jia, T. Tao, J. Sun, J. Du, Y. Xie, L. Yu, W. Tang, J. Wang and J. Gong, 1194 *Small Structures*, 2023, **4**, 2300139.
- 1195 124. G. Giordano, M. Carlotti and B. Mazzolai, *Adv. Mater. Technol.*, 2021, **6**, 1196 2100437.
- 1197 125. T. Cheng, Y. Z. Zhang, S. Wang, Y. L. Chen, S. Y. Gao, F. Wang, W. Y. Lai and W. Huang, *Adv. Funct. Mater.*, 2021, **31**, 2101303.
- 1199 126. Y. Zhang, X. Sun, Y. Ye, H. Oguzlu, Y. Zhu, J. Zhu, K. Le, P. Yang and F. 1200 Jiang, *Mater. Today*, 2024, **74**, 67-76.

- W. Li, S. Zheng, X. Zou, Y. Ren, Z. Liu, W. Peng, X. Wang, D. Liu, ZolSheng, View Article Online W. Li, S. Zheng, X. Zou, Y. Ren, Z. Liu, W. Peng, X. Wang, D. Liu, ZolSheng, View Article Online 1201 127.
- 1202 Y. Hu, J. Guo, Z. Sun and F. Yan, Adv. Funct. Mater., 2022, 32, 2207348.
- 1203 R. Ji, S. Yan, Z. Zhu, Y. Wang, D. He, K. Wang, D. Zhou, Q. Jia, X. Wang, B. 128.
- 1204 Zhang, C. Shi, T. Xu, R. Wang, R. Wang and Y. Zhou, Adv. Sci., 2024, 11, 1205 2401869.
- 1206 129. J. Yang, Z. Zhang, P. Zhou, Y. Zhang, Y. Liu, Y. Xu, Y. Gu, S. Qin, H. Haick 1207 and Y. Wang, *Nanoscale*, 2023, **15**, 3051-3078.
- 1208 Y. Wang, Soft Sci, 2024, 4. 130.
- 1209 Y. Wang, H. Haick, S. Guo, C. Wang, S. Lee, T. Yokota and T. Someya, Chem. 131. 1210 Soc. Rev., 2022, 51, 3759-3793.
- 1211 132. S. Cheng, Z. Lou, L. Zhang, H. Guo, Z. Wang, C. Guo, K. Fukuda, S. Ma, G. 1212
- Wang, T. Someya, H.-M. Cheng and X. Xu, Adv. Mater., 2023, 35, 2206793. 1213 S. Wei, R. Yin, T. Tang, Y. Wu, Y. Liu, P. Wang, K. Wang, M. Mei, R. Zou 133.
- 1214 and X. Duan, ACS Nano, 2019, 13, 7920-7929.
- 1215 B. Khan, Z. Riaz and B. L. Khoo, *Mater. Sci. Eng. R Rep.*, 2024, **159**, 100804. 134.
- 1216 P. A. Moreno-Sánchez, G. García-Isla, V. D. Corino, A. Vehkaoja, K. Brukamp, 135. 1217 M. Van Gils and L. Mainardi, Comput. Biol. Med., 2024, 108235.
- 1218 J. C. Hwang, M. Kim, S. Kim, H. Seo, S. An, E. H. Jang, S. Y. Han, M. J. Kim, 136. 1219 N. K. Kim and S.-W. Cho, *Sci. Adv.*, 2022, **8**, eabq0897.
- 1220 137. S.-H. Sunwoo, S. I. Han, C. S. Park, J. H. Kim, J. S. Georgiou, S.-P. Lee, D.-H. 1221 Kim and T. Hyeon, *Nat. Rev. Bioeng.*, 2024, **2**, 8-24.
- 1222 B. Pan, F. Xiong, J. Wang, J. Fu, Y. Ding, R. Qin and S. Li, Talanta, 2025, 138. 1223 127591.
- 1224 Y. Du, J. H. Kim, H. Kong, A. A. Li, M. L. Jin, D. H. Kim and Y. Wang, Adv. 139. Healthcare Mater., 2024, 13, 2303461. 1225
- 1226 140. S. Tang, D. Sha, Z. He, X. Chen, Y. Ma, C. Liu and Y. Yuan, Adv. Healthcare 1227 Mater., 2023, 12, 2300475.
- 1228 141. Y. Zhang, Q. Tang, J. Zhou, C. Zhao, J. Li and H. Wang, ACS Biomater. Sci. 1229 Eng., 2023, 10, 191-218.
- 1230 X. Shi, H. Yu, Z. Tang, S. Lu, M. You, H. Yin and Q. Chen, Sci. China Technol. 142. 1231 Sci., 2024, 67, 3136-3151.
- 1232 G. Yang, Z. Lan, H. Gong, J. Wen, B. Pang, Y. Qiu, Y. Zhang, W. Guo, T. Bu, 143. 1233 B. Xie and H. Wu, Adv. Funct. Mater., 2025, 35, 2417841.
- 1234 S. Chen, Q. Ouyang, X. Meng, Y. Yang, C. Li, X. Miao, Z. Chen, G. Zhao, Y. 144. 1235
- Lei, B. Ghanem, S. Gautam, J. Cheng and Z. Yan, Sci. Adv., 2025, 11, eadv2406. D. Farina, R. Merletti and R. M. Enoka, J. Appl. Physiol., 2004, 96, 1486-1495. 1236 145.
- 1237 146. H. Wang, Q. Ding, Y. Luo, Z. Wu, J. Yu, H. Chen, Y. Zhou, H. Zhang, K. Tao 1238 and X. Chen, Adv. Mater., 2024, 36, 2309868.
- 1239 S. Yang, J. Cheng, J. Shang, C. Hang, J. Qi, L. Zhong, Q. Rao, L. He, C. Liu 147. 1240 and L. Ding, Nat. Commun., 2023, 14, 6494.
- J. Wu, J. Xian, C. He, H. Lin, J. Li and F. Li, Adv. Mater., 2024, 36, 2405372. 1241 148.
- Y. Zhang, L. Chen, M. Xie, Z. Zhan, D. Yang, P. Cheng, H. Duan, Q. Ge and 1242 149.

View Article Online

DOI: 10.1039/D5TC01896J

- Open Access Article. Published on 05 2025. Downloaded on 29/10/2025 01:38:06.

 No This article is licensed under a Creative Commons Attribution-NonCommercial 3.0 Unported Licence.
- 1243 Z. Wang, *Mater. Today Phys.*, 2022, **27**, 100794.
- 1244 150. J. Park, Y. Lee, S. Cho, A. Choe, J. Yeom, Y. G. Ro, J. Kim, D.-h. Kang, S. Lee
- and H. Ko, Chem. Rev., 2024, 124, 1464-1534.
- 1246 151. H. Yuk, J. Wu and X. Zhao, *Nature Reviews Materials*, 2022, 7, 935-952.
- 1247 152. J.-W. Lee, M.-J. Shin, M.-H. Jang, W.-B. Jeong and S.-J. Ahn, *Med. Eng. Phys.*, 1248 2021, **98**, 65-72.
- 1249 153. V. Alcan and M. Zinnuroğlu, *Turkish journal of medical sciences*, 2023, **53**, 1250 1019-1031.
- 1251 154. Q. Liu, X. Xu, Y. Zhang, L. Liang, B. Zhang and S. Chen, *Chem. Eng. J.*, 2025, 1252 **509**, 161207.
- 1253 155. S. Roubert Martinez, P. Le Floch, J. Liu and R. D. Howe, *Adv. Healthcare* 1254 *Mater.*, 2023, **12**, 2202661.
- 1255 156. R. Wan, J. Yu, Z. Quan, H. Ma, J. Li, F. Tian, W. Wang, Y. Sun, J. Liu and D.
 1256 Gao, Chem. Eng. J., 2024, 490, 151454.
- 1257 157. R. Wan, S. Liu, Z. Li, G. Li, H. Li, J. Li, J. Xu and X. Liu, *J. Colloid Interface* 1258 *Sci.*, 2025, **677**, 198-207.
- 1259 158. T. Li, H. Qi, C. Zhao, Z. Li, W. Zhou, G. Li, H. Zhuo and W. Zhai, *Nat. Commun.*, 2025, **16**, 88.
- 1261 159. J. Gavvala, N. Abend, S. LaRoche, C. Hahn, S. T. Herman, J. Claassen, M.
- 1262 Macken, S. Schuele, E. Gerard and C. C. E. M. R. Consortium, *Epilepsia*, 2014, **55**, 1864-1871.
- 1264 160. C. De Gans, P. Burger, E. Van den Ende, J. Hermanides, P. Nanayakkara, R. 1265 Gemke, F. Rutters and D. Stenvers, *Sleep Med. Rev.*, 2024, 101951.
- 1266 161. M. Mohamed, N. Mohamed and J. G. Kim, *Biosensors*, 2023, **13**, 1019.
- 1267 162. J.-C. Hsieh, W. He, D. Venkatraghavan, V. B. Koptelova, Z. J. Ahmad, I.
- Pyatnitskiy, W. Wang, J. Jeong, K. K. W. Tang and C. Harmeier, *Device*, 2024, 2.
- 1270 163. X. Li, Y. Zhang, P. Tiwari, D. Song, B. Hu, M. Yang, Z. Zhao, N. Kumar and
 1271 P. Marttinen, *ACM Computing Surveys*, 2022, 55, 1-57.
- 1272 164. G. Yang, K. Zhu, W. Guo, D. Wu, X. Quan, X. Huang, S. Liu, Y. Li, H. Fang and Y. Qiu, *Adv. Funct. Mater.*, 2022, **32**, 2200457.
- 1274 165. M. Hu, J. Ren, Y. Pan, L. Cheng, X. Xu, C. L. Tan, H. Sun, Y. Shi and S. Yan,
 1275 Adv. Funct. Mater., 2024, 34, 2407926.
- 1276 166. G. Li, Y. Liu, Y. Chen, M. Li, J. Song, K. Li, Y. Zhang, L. Hu, X. Qi and X. 1277 Wan, *J. Neural Eng.*, 2023, **20**, 026017.
- 1278 167. J. Liu, S. Lin, W. Li, Y. Zhao, D. Liu, Z. He, D. Wang, M. Lei, B. Hong and H.
 1279 Wu, Research, 2022.
- 1280 168. W. U. Khan, Z. Shen, S. M. Mugo, H. Wang and Q. Zhang, *Chem. Soc. Rev.*, 1281 2025, **54**, 2832-2880.
- 1282 169. Q. Han, C. Zhang, T. Guo, Y. Tian, W. Song, J. Lei, Q. Li, A. Wang, M. Zhang and S. Bai, *Adv. Mater.*, 2023, **35**, 2209606.
- 1284 170. C. Wang, H. Wang, B. Wang, H. Miyata, Y. Wang, M. O. G. Nayeem, J. J. Kim,

- S. Lee, T. Yokota, H. Onodera and T. Someya, *Sci. Adv.*, 2022, **8**, eabol396_{1039/D5TC01896J}
- 1286 171. L. Li, X. Ye, Z. Ji, M. Zheng, S. Lin, M. Wang, J. Yang, P. Zhou, Z. Zhang, B.
- 1287 Wang, H. Wang and Y. Wang, *Small*, 2025, **21**, 2407996.
- 1288 172. G. Fang, X. Yang, Q. Wang, A. Zhang and B. Tang, *Materials Science and Engineering: C*, 2021, **127**, 112212.
- 1290 173. R. Lev and D. Seliktar, *J. Royal Soc. Interface*, 2018, **15**, 20170380.
- 1291 174. N. A. Alba, R. J. Sclabassi, M. Sun and X. T. Cui, *IEEE Trans. Neural Syst.*
- 1292 Rehabil. Eng., 2010, 18, 415-423.
- 1293 175. J. S. Blasco, E. Iánez, A. Ubeda and J. M. Azorín, *Expert Systems with*1294 *Applications*, 2012, **39**, 7908-7918.
- 1295 176. G. Perale, F. Rossi, E. Sundstrom, S. Bacchiega, M. Masi, G. Forloni and P. Veglianese, *ACS Chem. Neurosci.*, 2011, **2**, 336-345.
- 1297 177. K. Pradhan, G. Das, J. Khan, V. Gupta, S. Barman, A. Adak and S. Ghosh, 1298 *ACS Chem. Neurosci.*, 2018, **10**, 1535-1543.
- 1299 178. V. Martínez-Cagigal, J. Thielen, E. Santamaria-Vazquez, S. Pérez-Velasco, P. 1300 Desain and R. Hornero, *J. Neural Eng.*, 2021, **18**, 061002.
- 1301 179. J. Sosulski and M. Tangermann, J. Neural Eng., 2022, 19, 066001.
- 1302 180. M. Zheng, L. Li, X. Ye, Z. Ji, Y. Wang, Z. Wang, S. Lin, M. Wang, W. Yan, J.
- 1303 Yang, P. Zhou, Y. Zhang, R. Niu, H. Haick and Y. Wang, *Chem. Eng. J.*, 2025, 1304 **512**, 162451.
- 1305 181. X. Pan, J. Guan, S. Cao, X. Ma, Y. Ni and Q. Wang, *J. Colloid Interface Sci.*, 1306 2025, **680**, 753-761.
- 1307

View Article Online DOI: 10.1039/D5TC01896J

Data availability

The data that support the findings of this study are available from the corresponding author upon reasonable request.