A stretchable and adhesive ionic conductor based on polyacrylic acid and deep eutectic solvents

Gang Li1,5, Zhihao Deng1,5, Minkun Cai1, Kaixi Huang1, Mengxue Guo1, Ping Zhang6, Xingyu Hou1, Yuan Zhang1, Yueji Wang1, Yan Wang1, Xiang Wu3 and Chuan Fei Guo1,4,5

Hydrogels are a widely used ionic conductor in on-skin electronic and iontronic devices. However, hydrogels dehydrate in the open air and freeze at low temperatures, limiting their real applications when they are attached on skin or exposed to low temperatures. Polymer-liquid ionic gels can overcome these two obstacles, but synthetic liquid ionic gels are expensive and toxic. In this work, we present an ionic conductor based on polyacrylic acid (PAAc) and deep eutectic solvents (DESs) that well addresses the aforementioned challenges. We polymerize acrylic acid in DESs to get the PAAc–DES gel, which exhibits excellent stretchability (>1000%), high electrical conductivity (1.26 mS cm−1), high adhesion to the skin (~100 N m−1), as well as good anti-drying and anti-freezing properties. We also demonstrate that the PAAc–DES gel can be used as an on-skin electrode to record the surface electromyographic signal with high signal quality, or as a transparent stretchable electrode in iontronic devices that can work at ~20 °C. We believe that the PAAc–DES gels are an ideal candidate as epidermal electrodes or transparent stretchable electrodes.

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INTRODUCTION

Soft ionic conductors are a vital component in on-skin electronics that can be used as an interface to transfer electrical signals between the human body and the external devices1–5. A desired ionic conductor should exhibit high electrical conductance, high stretchability, as well as low cost for commercialization. Hydrogels are a promising material to conduct ions6–9, exhibiting skin-like mechanical performance, good biocompatibility, high electrical conductivity, and other attractive properties10–13. Therefore, hydrogels have been applied in electrophysiological signal recording1, human–machine interaction14,15, ionic skins16, and luminescent devices17,18. However, the properties of hydrogels are highly related to water content, while hydrogels dehydrate rapidly in open air. A dehydrated hydrogel loses its softness, stretchability and changes its shape. Therefore, additional seals or chemicals that prevent dehydration are often needed to prevent water loss19. In addition, hydrogels tend to freeze at a low temperature, which may hinder their applications in cold weather.

Polymer–ionic liquid gels (IL gels) are another selection of soft ionic conductors. Ionic liquids (ILs) are salts of low melting point, and they are non-volatile at room temperature15,16, eliminating the problem of easy freezing and dehydration in hydrogels. As a result, the IL gels exhibit higher stability than the hydrogel. IL gels have demonstrated usefulness in various ionic devices17–19. However, synthetic ILs are expensive and toxic, being major problems for the commercialization and on-skin applications of the IL gels20.

Deep eutectic solvents (DESs) are an emerging alternative to ILs, which are formed by mixing a quaternary ammonium salt (e.g., choline chloride, ChCl) with another hydrogen bond donor (e.g., ethylene glycol, EG)21. DESs possess similar physico-chemical properties to those of common ILs, such as high electrical conductivity and low vapor pressure. The most pronounced advantage of DESs over ILs is their low cost and excellent biocompatibility22–24, and thus DES-based ionic conductors are rapidly developed. However, DES gels have not yet been comparable with common IL gels in terms of stretchability, transparency, and adhesive property25–27.

In this work, we show that a stretchable, transparent, adhesive, and anti-freezing DES gel can be prepared by polymerizing monomers in DESs, by mixing acrylic acid (AAC) with various DESs and initiating the reaction via ultraviolet (UV) radiation, similar to the method to make synthetic hydrogels. The prepared PAAc–DES gel is anti-drying and anti-freezing, demonstrating higher stability than the hydrogels. The gel also exhibits a high transparency of ~92%, a large stretchability over 1000%, at moderate electrical conductivity of 1.26 mS cm−1, and strong adhesion (~100 N m−1) with the human skin, which are comparable to most IL gels. The cost of the PAAc–DES gels, however, is about two orders of magnitudes lower than that of many synthetic ILs. The PAAc–DES gels also exhibit high compatibility on human skin without showing skin irritation after wearing for 8 h. Thus, this soft ionic conductor has been applied as an on-skin electrode to record bioelectricity signals, exhibiting low impedance of the skin–electrode interface and improved signal quality compared with commercial electrodes. The anti-freezing PAAc–DES gel also can serve as an ionic skin working at ~20 °C. We believe that our work can enrich the ionic conductors with a cheaper, eco-friendly, and high-performance choice.

RESULTS

Preparation and basic properties of PAAc–DES gel

We mixed ChCl with four hydrogen bond donors including glycerol (Gly), urea, diethylene glycol (DG), and ethylene glycol (EG)21. DESs possess similar physico-chemical properties to those of common ILs, such as high electrical conductivity and low vapor pressure. The most profound advantage of DESs over ILs is their low cost and excellent biocompatibility22–24, and thus DES-based ionic conductors are rapidly developed. However, DES gels have not yet been comparable with common IL gels in terms of stretchability, transparency, and adhesive property25–27.

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crosslinker poly(ethylene glycol) diacrylate (PEDGA), and initiator 2-oxoglutaric acid in DESs. The chemical structures of the molecules are shown in Fig. 1a. The precursor solution comprises 28.9 wt.% AAc and 70 wt.% DES, and the weight percentage of the crosslinker and initiator is 0.1% and 1%, respectively. We clarify the four PAAc–DES gels according to their composition, such as PAAc–ChCl–Gly gel in this text. The AAc is dissolved in all DESs to form clear solutions and all solutions are polymerized into a transparent gel (Fig. S1), indicating the high compatibility between the PAAc and the DESs. The UV–Vis spectra in Fig. 1b show that the PAAc–ChCl–EG gel has the highest transparency (~92%), whereas the PAAc–ChCl–Urea gel has the lowest conductivity (0.05 mS cm⁻¹). Therefore, we chose the PAAc–ChCl–EG gel as the representative of the PAAc–DES gels to conduct further study for bio-electrical signal measurement and application in iontronic devices.

**Cyclic stretch-release and dynamic test**

The PAAc–ChCl–EG gel exhibits good recovery ability when it is released from elongation. Samples are subjected to loading–unloading tests at different strains, and the stress–strain curves are shown in Fig. 2a. The initial residual strain of the gel after releasing from 100% strain is about 20% and increases to 40% when the gel is stretched to 400%. These samples are allowed to recover for 1 min and then subjected to a second stretch–release cycle. The stress–strain curves are shown in Fig. S3a–d, which indicate that the majority of the initial residual strain is removed. The sample is then tested over 100 cycles of loading–unloading, and the selected stress–strain curves are shown in Fig. S4. After 100 cycles of stretch–release, the stress at 100% strain reduces to ~60% of its initial value, indicating that internal structures are partially broken. After 10 min, the strength of the PAAc–ChCl–EG gel and the highest tensile strength (296 ± 21 kPa). By comparison, the modulus of the PAAc–ChCl–DG gel is only 4 kPa, which is the softest one. The other two PAAc–DES gels have similar moduli of ~20 kPa. All gels show moderate tensile strengths > 100 kPa. We also measured the ionic conductivity of these gels at room temperature. The Nyquist plots and the calculated ionic conductivities are given in Fig. 1d, e, respectively. The PAAc–ChCl–EG gel has the highest conductivity (1.26 ± 0.13 mS cm⁻¹), whereas the PAAc–ChCl–Urea gel has the lowest conductivity (0.05 mS cm⁻¹). Therefore, we chose the PAAc–ChCl–EG gel as the representative of the PAAc–DES gels to conduct further study for bio-electrical signal measurement and application in iontronic devices.
of the gel restores to ~75% of its initial value and the residual strain reduces to 4%. Because the ionic conductor serves as a stretchable electrode, the important consideration is the dimension stability instead of strength. In this sense, the PAAc–ChCl–EG gel performs well because it has a small residual strain after being elongated to large strains or being stretched for 100 cycles, enabling possible applications as a stretchable electrode.

The dynamic mechanical properties of the PAAc–ChCl–EG gel at different temperatures are shown in Fig. 2b, which exhibit frequency-dependent storage modulus ($G'$) and loss modulus ($G''$) in the temperature range of −30−50 °C. The storage modulus becomes larger as the oscillating frequency increases. For instance, at room temperature, $G'$ of the gel is 10.6 kPa at 0.1 Hz, and it reaches 368.7 kPa at 100 Hz. This is because that the gel contains high solid content, and the entanglement of polymer chains contributes more to the $G'$ at a shorter time scale, or higher frequency. Another observation is that the $G'$ at 0.1 Hz of the gel only increases to 18.6 kPa at −30 °C compared with the one measured at 25 °C. That means, the gel becomes two times harder after it drops from room temperature to an ultralow temperature of −30 °C, at which most hydrogels freeze to have a rigidity increase of ~5 orders of magnitude.

**Thermostability**

We tested the thermal stability of the gel using thermogravimetric analysis (TGA), and the result is shown in Fig. 2c. The PAAc–ChCl–EG gel loses only ~4% of its initial mass at 100 °C, indicating that the gel can be used at a relatively high temperature. For comparison, we prepared a PAAc hydrogel containing the same content of PAAc and conducted the same test, as shown in Fig. 2c. The hydrogel starts to lose water as the test begins, and the mass of the hydrogel reduces to 70% when the temperature reaches 100 °C. We measured the weight change of the PAAc–ChCl–EG gel and the PAAc hydrogel at 37 °C for 24 h and it is plotted in Fig. 2d. The hydrogel loses 57% of the water within 2 h and has a weight loss of ~60% over 24 h. The evaporation of water from the hydrogel results in significant shrinkage in dimensions, as shown in Fig. S5. In contrast, the weight of the PAAc–ChCl–EG gel maintains stability throughout the test. In the first few hours, the weight change of the PAAc–ChCl–EG gel is about 1%, and after 24 h, the PAAc–ChCl–EG gel loses ~3% of its mass due to the slow evaporation of EG. These results indicate a far higher thermostability of the PAAc–DES gel than hydrogels.

**Comparison of the PAAc–DES gel with other IL gels and DES gels**

The performances of the PAAc–DES gel are compared with those of other representative non–water ionic conductors in Table 1. Our PAAc–DES gel is comparable to other materials in terms of transparency, but one order of magnitude higher than that of other DES gels in stretchability ($\lambda$). In addition, the stretchability and electrical conductivity of our PAAc–DES gel are comparable to that of most IL gels. Price is another great concern in practical applications, and our PAAc–DES gel is superior in this consideration due to the low price of the chemicals used. The price is < $10 kg$⁻¹ for ChCl and < $1 kg$⁻¹ for EG, which are several orders of magnitude lower than that of most synthetic ionic liquids (Table S1). In this sense, our DES gels are a better choice than IL gels since they present a similar performance with ILs but have a much lower cost.

**Adhesion of the PAAc–DES gel and its application as an on-skin electrode**

The PAAc–ChCl–EG can adhere to various substrates, such as metals and the human skin. Figure. 3a shows that a piece of PAAc–ChCl–EG that is adhered to two copper strips. When we pulled the two copper strips away to a large tensile strain up to 800%, the PAAc–DES gel–copper interface is tough enough to prevent the separation of the two materials. We measured the
interfacial toughness by 180° peeling, as shown in Fig. 3b and Fig. S6. The toughness values between PAAc-ChCl-EG gel and copper, steel, and tin are 109 ± 12 N m⁻¹, 129 ± 10 N m⁻¹, and 85 ± 9 N m⁻¹, respectively. The gel is also highly adhesive to the human skin. Fig. 3c shows that when the free end of a gel sample undergoes large deformation, the other end is still well adhered to the skin and causes large skin deformation. In Fig. 3d, we compare the adhesion performance of the PAAc-ChCl-EG gel with that of commercial 3M VHB tapes. The interfacial toughness between our gel and the skin is 173 kΩ, which is larger than that of the skin–VHB tape interface (< 70 N m⁻¹). The strong adhesion of the PAAc-DES gel is originated from the abundant –COOH function groups of PAAc, which interact with the adherend by Van der Waals force. Another contribution to the strong adhesion is the viscoelasticity of the PAAc-ChCl-EG gel achieved by tuning the crosslinking density. This can be evidenced in Fig. S7, showing that both the G' and G" of the PAAc-ChCl-EG gel are strongly dependent on the density of crosslinking. If no crosslinker is added, the linear PAAc and DES form a sticky product and a residue will be left on the skin after removing the gel. By contrast, if crosslinking density is too high, the gel is no longer adhesive, and the residue will be left on the skin after removing the gel. By contrast, if crosslinking density is too high, the gel is no longer adhesive, and the residue will be left on the skin after removing the gel. By contrast, if crosslinking density is too high, the gel is no longer adhesive, and the residue will be left on the skin after removing the gel.

We used our PAAc-ChCl-EG gel as surface electrodes and compared our gel with commercial Ag–AgCl electrodes serving as control samples. The PAAc-ChCl-EG gel was first cut in the same dimensions as the commercial Ag–AgCl electrodes and then adhered to both our electrodes and the control samples were attached to the biceps brachii. The impedance values (|Z|) of both electrodes were measured and the results are provided in Fig. 3g. Over most of the frequency range, our gel has a lower impedance than that of the commercial one. For instance, the |Z| of the DES gel is 173 kΩ at 10 Hz and 4.4 kΘ at 1 kHz, while those for the commercial electrode are 238 kΘ and 123 kΘ, respectively. The low impedance lies in the high electrical conductance of our gel and the conformal contact between the PAAc-ChCl-EG gel and skin. Electromyographic (EMG) signals recorded using the DES gel electrodes and commercial electrodes are shown in Fig. 3h. The signals are generated from the same action of squeezing a hand gripper by the same person. The amplitudes of both EMG signals are almost the same, but the one recorded from our electrodes has a lower noise level than that recorded from the Ag–AgCl electrodes (Fig. S9). The improved EMG signal quality is believed to be related to the lower impedance of the PAAc–DES gels, which might be extended to the collection of other bioelectricity signals.

**Anti-freezing ionic skin**

The PAAc-ChCl-EG gel possesses a high anti-freezing performance, which allows the gel to work under low temperatures down to −60 °C. Fig. 4a presents the differential scanning calorimetric (DSC) curve of the PAAc-ChCl-EG gel that is cooling from 20 °C to −60 °C, showing a flat curve that indicates no glass transition in this temperature range. For comparison, an exothermic peak located near −15 °C has appeared in the DSC curve of the PAAc hydrogel, indicating that the hydrogel begins to freeze at this temperature. In Fig. S10, we show photographs of the PAAc hydrogel and the PAAc-DES gel kept at −60 °C. The hydrogel is frozen into a white and stiff solid, whereas the PAAc-ChCl-EG gel remains transparent. We pulled both specimens at −60 °C, and the frozen hydrogel lost its stretchability, while the PAAc-ChCl-EG gel remained soft and stretchable. We thus anticipate that the PAAc-ChCl-EG gel can be used in extremely low-temperature environments, including the Antarctic Pole. Besides, it is worth noting that the glass transition temperature of the PAAc-ChCl-EG gel is lower than that of the pure PAAc (~130 °C, see Fig. S11) and the freezing temperature of ChCl-EG (~36 °C). This anti-freezing behavior may be attributed to the interaction between the PAAc and the ChCl–EG, in which the PAAc hinders the crystallization of the solvent and the solvent also softens the polymer chains. After all, this observation needs further work to provide a valid explanation.

The high stretchability of the PAAc-DES gel under low temperatures allows for applications in anti-freezing ionic skin. The device structure of the ionic skin is illustrated in Fig. 4b, with two DES gel electrodes sandwiching an insulating VHB tape serving as the dielectric. A photograph of this ionic skin is shown in Fig. S12, in which the gel is dyed for better distinction. The sensitivity of the ionic skin under pressing is 5 MPa⁻¹, and the gauge factor under stretch is 0.42 (Fig. 4c, d); both are comparable to the previous results. Upon pressing, the sensor can respond to the stimuli both at room temperature and at −20 °C, as shown in Fig. 4e, f, suggesting that the ionic skin can work at −20 °C. However, the signal intensity at −20 °C is weaker than that at room temperature because of the increased rigidity of the gel at a lower temperature. For other hydrogel-based ionic skins, the whole device will lose sensing function at such a low temperature. We also demonstrated that the PAAc-ChCl-EG gel can be used in anti-freezing ionotropic luminescence that works at a temperature lower than −16 °C (Fig. S13). Low-temperature tolerance is vital for on-skin electronics, especially in outdoor applications. Here our results indicate that the PAAc–DES gel is a promising electrode for anti-freezing iontronics under extremely low-temperature environments.

**DISCUSSION**

In this work, we demonstrated the great potential of the PAAc–DES gel in the field of on-skin electrodes and iontronics devices. The PAAc–DES gel is fabricated by UV curing AAC in DES. The obtained PAAc–DES gels show high stretchability, transparency, and adhesive performance that are comparable with hydrogels and IL gels. Our PAAc–DES gels present the advantage of non-volatility and anti-freezing ability, as well as low price. Such advantages make the PAAc–DES gels a superior alternative to IL

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**Table 1.** Comparison of our work with representative IL gels and DES gels.

| Type   | Composition (polymer and solvent) | T (%) | λ (%) | θ (mS cm⁻¹) | Price | Ref. |
|--------|----------------------------------|-------|-------|-------------|-------|------|
| IL gel | PAAc and [EMIM] [DCA]            | > 85  | 850   | 19          | Expensive | 17   |
|        | PEA and [BMMim][TFSI]           | 93.6  | > 1000| 0.01–1      | Expensive | 18   |
|        | PAAc & PDMAPS and [EMIM] [ES]   | > 90  | > 10 000| ~ 0.1      | Expensive | 19   |
|        | P(VDF-HFP) & P(MMA-BMA) and [EMIM][TFSI] | 93   | 307   | > 1        | Expensive | 28   |
| DES gel| Gelatin and [ChCl][EG]           | 80–90 | > 300 | 2.5         | Very cheap | 25   |
|        | PAAm and [ChCl][urea, glycerol]  | –     | > 200 | ~ 1        | Very cheap | 26   |
|        | PAAc and [ChCl][EG]             | 92    | 1100  | 1.26       | Very cheap | This work |
gels. We have shown that our gel can be used as an on-skin electrode to record surface EMG signals and to construct ionic skins that can sense external stimuli at low temperatures down to –20 °C. Such low-temperature applications of the PAAc–DES gels may be extended to other devices not demonstrated in this work. We believe that the PAAc–DES gels are a promising alternative to the hydrogels and IL gels in various circumstances.

**METHODS**

**Materials**

Choline chloride (ChCl), glycerol (Gly), diethylene glycol (DG), ethylene glycol (EG), urea, acrylic acid (AAc), poly(ethylene glycol) diacrylate (PEGDA, Mw. 600), and 2-oxoglutaric acid were purchased from Shanghai Aladdin Biochemical Technology Co., Ltd. Silicon rubber (Sylgard 184) was
Preparation of PAAc–DES gel

The DESs were prepared by mixing ChCl with hydrogen bond donors, including Gly, urea, DG, and EG, in a molar ratio of 1:2. The mixtures were then stirred at 100 °C for 2 h. Next, 2.89 g AAc, 7 g DES, 0.1 g 2-oxoglutaric acid, and 0.01 g PEGDA were added into a vial, and VORTEX 3 was used to accelerate dissolution. The PAAc–DES precursor was injected into a glass mold separated by spacers, and then the glass mold was placed under a 365 nm UV radiation for 3 min. A PAAc hydrogel was prepared using the same steps by changing DES to water.

Basic properties characterization

The UV–Vis spectra of the PAAc–DES gels were recorded by the PE LAMBDA 950 instrument in the range of 400–800 nm. The thickness of the sample was 1 mm, and the air was taken as a reference.

The tensile properties of the PAAc–DES gels were measured by a universal testing machine (XLD-20E, Jingkong Mechanical testing Co., Ltd). The specimens were cut into a dumbbell shape (35 mm in total length, 2 mm in inner width, and 12 mm in gauge length) and stretched at a speed of 100 mm min⁻¹.

The bulk resistance $R_b$ of the PAAc–DES gel was measured by the AC impedance spectroscopy measurements (CS series, Wuhan CorrTest Instruments Corp., Ltd.). The ionic conductivities $\sigma$ of the PAAc–DES gels were calculated by the formula $\sigma = L/S\Delta C/\Delta t$, where $L$ and $S$ are the thickness and cross-section area of the specimen, respectively.

The rheology property of the PAAc–ChCl–EG gel was measured by a HAAKE MARS III rheometer using a 20 mm parallel plate geometry. Frequency sweep was measured at a shear strain of 1% at different temperatures.

The thermostability of the PAAc–ChCl–EG gel and PAAc hydrogel was measured by a thermogravimetric analyzer (Discovery TGA) at a heating rate of 10 K min⁻¹ under nitrogen atmosphere.

The weight change of the PAAc–ChCl–EG gel and PAAc hydrogel at 37 °C was recorded at different time intervals; after the weighing, the samples were stored in an oven at a pre-set temperature.

The thermal behavior of the PAAc–ChCl–EG gel and PAAc hydrogel were studied by differential scanning calorimetry (DSC). About 5 mg of the PAAc–ChCl–EG gel and PAAc hydrogel were put in the pan and heated or cooled at a speed of 10 K min⁻¹ in nitrogen in a Discovery DSC.
The 90° and 180° peeling tests were conducted by cutting the specimen are cut into a dimension of 100 × 10 × 1 mm³ and glued to a PI tape as stiff backing to prevent elongation during testing. The test speed was 50 mm min⁻¹.

**Impedance measurement and EMG recording**

A pair of the electrodes (the PAAc-ChCl-EG gel or the Ag–AgCl gel electrodes (CH508R, Nison Instrument (Shanghai Limited)) adhered on *biceps brachii* separated by a distance of 2 cm. The impedance was measured by an electrochemical workstation (CS series, Wuhan ConTest Instruments Corp., Ltd.) from 1 Hz to 10 kHz. Electromyographic (EMG) signals were recorded using the micro-4 system (Cambridge Electronic Design Limited) in a frequency range from 1 to 1000 Hz (50 Hz is filtered). Two working electrodes were attached to the same position for impedance measurement; a reference electrode was placed at the elbow. The action was squeezing a hand gripper.

**Ionic skin fabrication and measurement**

Two pieces of PAAc–ChCl–EG gels were adhered to both sides of a 3M VHB tape; one end of the PAAc–ChCl–EG gel was connected to a silver wire. The capacitance signal was obtained from a Keysight E4980AL LCR meter, the working frequency was 10 kHz.

**Fabrication of ionotropic luminescence**

ZnS:Cu particles were mixed with the PDMS base and curing agent in a weight ratio of 1:10.1. The mixture was spin coated on a piece of glass at 300 rpm for 1 min, and then heated at 65 °C for 4 h, forming the phosphor layer. The device was fabricated by sandwiching the phosphor layer between two pieces of PAAc–ChCl–EG gel. Copper foils were attached onto the DES gel to connect the power source (Trek 615). The cold environment is provided by a cold trap and the temperature is measured by using an Infrared camera (226 s, Fotric, USA).

**Experiments on human subjects**

All experiments were conducted under approval from the Institutional Review Board at the Southern University of Science and Technology (protocol number: 20190007).

**Reporting summary**

Further information on research design is available in the Nature Research Reporting Summary linked to this article.

**DATA AVAILABILITY**

The experimental data referenced in this text are available from the authors upon reasonable request.

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**REFERENCES**

1. Pan, L. et al. A compliant ionic adhesive electrode with ultralow bioelectronic impedance. *Adv. Mater.* **32**, 2003723 (2020).
2. Yuk, H., Lu, B. & Zhao, X. Hydrogel bioelectronics. *Chem. Soc. Rev.* **48**, 1642–1667 (2019).
3. Liu, X., Liu, J., Lin, S. & Zhao, X. Hydrogel machines. *Mater. Today* **36**, 102–124 (2020).
4. Lopes, P. A. et al. Soft bioelectronic stickers: selection and evaluation of skin-interfacing electrodes. *Adv. Healthc. Mater.* **8**, 1900234 (2019).
5. Wu, H. et al. Materials, devices, and systems of on-skin electrodes for electro-physiological monitoring and human–machine interfaces. *Adv. Sci.* **8**, 2001938 (2021).
6. Rong, Q., Lei, W. & Liu, M. Conductive hydrogels as smart materials for flexible electronic devices. *Chem. Eur. J.* **24**, 16930–16943 (2018).
7. Yang, C. & Suo, Z. Hydrogel iontronics. *Nat. Rev. Mater.* **3**, 125–142 (2018).
8. Lee, Y., Song, W. J. & Sun, J. Y. Hydrogel soft robotics. *Mater. Today Phys.* **15**, 100258 (2020).
9. Zhang, D. et al. Self-powered ionic sensors overcoming the limitation of ionic conductors as wearable sensing devices. *Mater. Today Phys.* **15**, 100246 (2020).
10. Zhang, Q., Liu, X., Duan, L. & Gao, G. Nucleotide-driven skin-attachable hydrogels toward visual human–machine interfaces. *J. Mater. Chem. A.* **8**, 4515–4523 (2020).
11. Chen, L. et al. 3D printed super-anti-freezing self-adhesive human-machine interface. *Mater. Today Phys.* **19**, 100404 (2021).
12. Sun, J. Y., Keglinger, C., Whitesides, G. M. & Suo, Z. Ionic skin. *Adv. Mater.* **26**, 7608–7614 (2014).
13. Yang, C. et al. Ionotronic luminescent fibers, fabrics, and other configurations. *Adv. Mater.* **32**, 2005545 (2020).
14. Yang, C. H. et al. Electroluminescence of giant stretchability. *Adv. Mater.* **28**, 4480–4484 (2016).
15. Galiriusi, M., Lewandowski, A. & Stepien, I. Ionic liquids as electrolytes. *Electrochim. Acta.* **51**, 5567–5580 (2006).
16. Dong, K. et al. Multiscale studies on ionic liquids. *Chem. Rev.* **117**, 6636–6695 (2017).
17. Lai, J. et al. Highly stretchable, fatigue-resistant, electrically conductive, and temperature-tolerant ionogels for high-performance flexible sensors. *ACS Appl. Mater. Interfaces* **11**, 26412–26420 (2019).
18. Shi, L. et al. Highly stretchable and transparent ionic conductor with novel hydrophobicity and extreme-temperature tolerance. *Research 2020*, 2505619 (2020).
19. Lei, Z. & Wu, P. A highly transparent and ultra-stretchable conductor with stable conductivity during large deformation. *Nat. Commun.* **10**, 1–9 (2019).
20. Zhang, Q., Vigier, K. D. O., Royer, S. & Jerome, F. Deep eutectic solvents: syntheses, properties and applications. *Chem. Soc. Rev.* **41**, 7108–7146 (2012).
21. Paiva, A. et al. Natural deep eutectic solvents - solvents for the 21st century. *ACS Sustain. Chem. Eng.* **2**, 1063–1071 (2014).
22. Smith, E. L., Abbott, A. P. & Ryder, K. S. Deep eutectic solvents (DESs) and their applications. *Chem. Rev.* **114**, 11060–11082 (2014).
23. Macário, L. et al. Cytotoxicity profiling of deep eutectic solvents to human skin cells. *Sci. Rep.* **9**, 1–9 (2019).
24. Crump, M. R. et al. Sensorized tissue analogues enabled by a 3D-printed conductive organogel. *npj Flex. Electron.* **5**, 1–8 (2021).
25. Qin, H., Owyang, R. E., Sonkusale, S. R. & Panzer, M. J. Highly stretchable and nonvolatile gelatin-supported deep eutectic solvent gel electrolyte-based ionic skins for strain and pressure sensing. *J. Mater. Chem. C.* **7**, 601–608 (2019).
26. Hong, S. et al. A stretchable and compressible ion gel based on a deep eutectic solvent applied as a strain sensor and electrolyte for supercapacitors. *J. Mater. Chem. C.* **8**, 550–560 (2020).
27. Qin, H. & Panzer, M. J. Chemically cross-linked poly (2-hydroxyethyl methacrylate)-supported deep eutectic solvent gel electrolytes for eco-friendly super-capacitors. *ChemElectroChem* **4**, 2556–2562 (2017).
28. Gu, Z. et al. Skin adhesives with controlled adhesion by polymer chain mobility. *ACS Appl. Mater. Interfaces* **11**, 1496–1502 (2018).
29. Hansen, B. B. et al. Deep eutectic solvent: a review of fundamentals and applications. *Chem. Rev.* **121**, 1232–1285 (2021).

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**AUTHOR CONTRIBUTIONS**

G.L., Z.D., and C.J.F. designed the project and experiments. G.L. and Z.D. fabricated the materials and conducted the characterization. M.C. tested the performance of the ionic skin. P.Z. fabricated the ionotronic luminescence. All authors discussed the results and commented on the manuscript.

**COMPETING INTERESTS**

The authors declare no competing interests.

**ADDITIONAL INFORMATION**

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Correspondence and requests for materials should be addressed to C.F.G.

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