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Self-healing Ionic Liquid-based Electronics and Beyond

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Abstract Owing to the advantages of non-volatility, outstanding fluidity and easy recyclability, ionic liquid-based electronics, such as thermometer, strain sensors and thermoelectric converters, have been growing as attractive alternatives to traditionally solid electronics. The fluidic character endows the ionic liquid-based circuit with self-healing ability, satisfying the needs of longer lifetime and less waste generation for electronics, while at the same time brings the risk of leakage. Avoiding the leakage without sacrifice of self-healing ability is one of the major challenges for constructing ionic liquid-based electronic devices. In this feature article, we summarize our recent progresses in developing two types of self-healing electrical devices based on ionic liquids with little risk of leakage. One type involves the encapsulation of ionic liquids in self-healing polymers, and the other type uses ionic polymers or free-standing ionic liquids which are successfully formulated as intrinsically conductive, self-healing, and recyclable electronic devices without additional encapsulation. In the end, a comprehensive outlook is prospected for the future development of ionic liquid-based self-healing electronics, which is expected to spur more innovative work in this field.

Keywords Self-healing electronics; Self-healing materials; Recyclable electronics; Ionic liquid; Liquid leakage

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INTRODUCTION

As the booming development of the Internet-of-Things, we have witnessed an exponential rise in our dependence on electronics. However, the increasing number of electronic devices are leading to heavy environmental burdens after broken or being disposed.[1–4] Traditionally, solid semiconducting materials have always been among top-priority choices for building electronic devices, which yet possess limited flexibility and ductility against significant deformation. Once broken, the whole device would completely malfunction and then be disposed. On the other hand, the rapid renewal of mobile or wearable electronics extremely shortens the average life span of electronic devices, which would become a major branch of soil and water pollution after disposed without proper recycling procedures. These two challenging items have been puzzling researches all along with the innovation of new electrical materials and devices. In this context, the past decade witnessed a tremendous increase of research interests in the interdisciplinary cooperation between material science, engineering, and soft electronics, aiming for soft or green electronics. For example, pliable or stretchable electronic devices have been preferred for avoiding the vulnerability of traditional devices, presenting infinite potential in wearable electronic devices for daily health monitoring or implantable bioelectronic devices for *in vivo* biological detection.[5–23] However, it still fails to resist the unexpected mechanical damage led by the sudden tearing or repeated stretching.[24] Hence, these soft electronics are required with the ability of self-repairing after receiving mechanical damages, just like the human skins. Driven by this demand, self-healing materials[25–34] have been widely exploited and integrated with conductive or even sensing elements, intending to construct self-healing electronic devices.[35–43]

Unlike traditionally solid conductive or semiconductive materials, liquid conductive materials exhibit unique deformability and self-healing abilities, both of which are ideal parameters for soft electronic devices. For example, when encountering unexpected damages, the liquid conductive circuit can be instantly repaired *via* the merging of broken parts of liquid. Besides, with the benefit of doping capability, the function of liquid conductive materials can be expanded in addition to electrical applications. Among various liquid conductive materials, ionic liquid with intrinsic advantages of negligible volatility, electrochemical stability, high solubility, and terrific inflammability is one of the most promising candidates for the fabrication of liquid-based electronic devices.[44–50] In 2014, our group reported that the conductivities of imidazolium-based ionic liquids exhibited ultra-high sensitivity against temperature change, which has been inspiring significant interests in developing ionic liquid-based electronic devices that integrate with excellent sensitivity, flexibility, or even self-healing ability.[51,54–66] However, just like a double-edged sword, a key trade-off is that the intrinsic fluidity of ionic liquid also brings the risk of leakage, especially when the ionic liquid-based devices suffer from unexpected damages. Therefore, in order to push forward this new area of self-healing

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electronics based on ionic liquids, there is also an essential need for exploring effective strategies that can solve the leakage issue.

In the past few years, our group has put great efforts in integrating self-healing polymers or moieties with ionic liquids, aiming to constructing ionic liquid-based electronic devices with excellent self-healing ability or even recyclability with little risk of liquid leakage. Herein, in this feature article, we summarize those representative examples with the success of preventing liquid leakage during self-healing processes, which are mainly divided into two categories: (1) self-healing polymer-assisted ionic liquid-based electronics;[62,63] (2) intrinsically self-healing and recyclable ionic liquid-based electronics.[64–66] This feature article in the special issue focusing on self-healing polymers is expected to stimulate worldwide interests in self-healing electronics and to inspire more interdisciplinary researches focusing on the fundament and application of self-healing materials and ionic liquid-based electronics.

SELF-HEALING POLYMER-ASSISTED IONIC LIQUID-BASED ELECTRONICS

With respect to the fluidic characteristics of ionic liquids, appropriate sealing approaches are necessary for minimizing the risk of leakage during the service of ionic liquid-based electronic devices. For example, inspired by the water absorption effect of porous foam, we developed a pressure/temperature dual sensor based on ionic liquids partially filled in a polydimethylsiloxane (PDMS) foam.[51] This facile while effective approach illustrates that the capillary effect could offer substantially physical interactions for pinning or even sealing the ionic liquids.[52,53] Similarly, microfluidic chip that allows liquid to flow and functionalize in tiny microchannels is really another platform for sealing ionic liquids. However, in order to realize effective sealing without sacrificing the self-healing ability of ionic liquids, the sealing matrix is preferentially considered with the ability of self-healing for fabricating self-healing electronic devices.

From this viewpoint, a type of self-healing polymers (SHPs) was designed and synthesized *via* a two-step consecutive polycondensation reaction among the mixture of dicarboxylic and tricarboxylic fatty acid, diethylenetriamine (DETA), and urea.[62] As shown in Fig. 1(a), the SHPs formulated with large amount of amide and urea bond, as well as part of unreacted carboxylic or amine moiety, were mostly crosslinked by the strong hydrogen bonding among those hydrogen donors and acceptors. Hence, the SHPs exhibited desirable self-healing ability based on the quick reconfiguration of these hydrogen bonds, which could be remarkably repaired within 10 min even at room temperature (Fig. 1b). The repaired sample nearly persevered similar mechanical properties with its originated form, including tensile or bending behaviors. Since the hydrogen bonds are thermally sensitive, the healing rate could be accelerated *via* increasing the healing temperature. For instance, once the temperature was increased to 50 °C, the rejointed sample of two broken parts could completely recover to its original tensile performance in 5 min. Besides, owing to the thermally reversible crosslinking of hydrogen bonds, the SHPs could be adapted with thermal processing or even thermal casting, which afforded great convenience for fabricating microchannels with appropriate topologies to enclose ionic liquids.

Although both the polymer matrix and ionic liquid are self-repairable once rejointed from the fracture sites, the liquid leakage after the device fracture should be avoided by con-

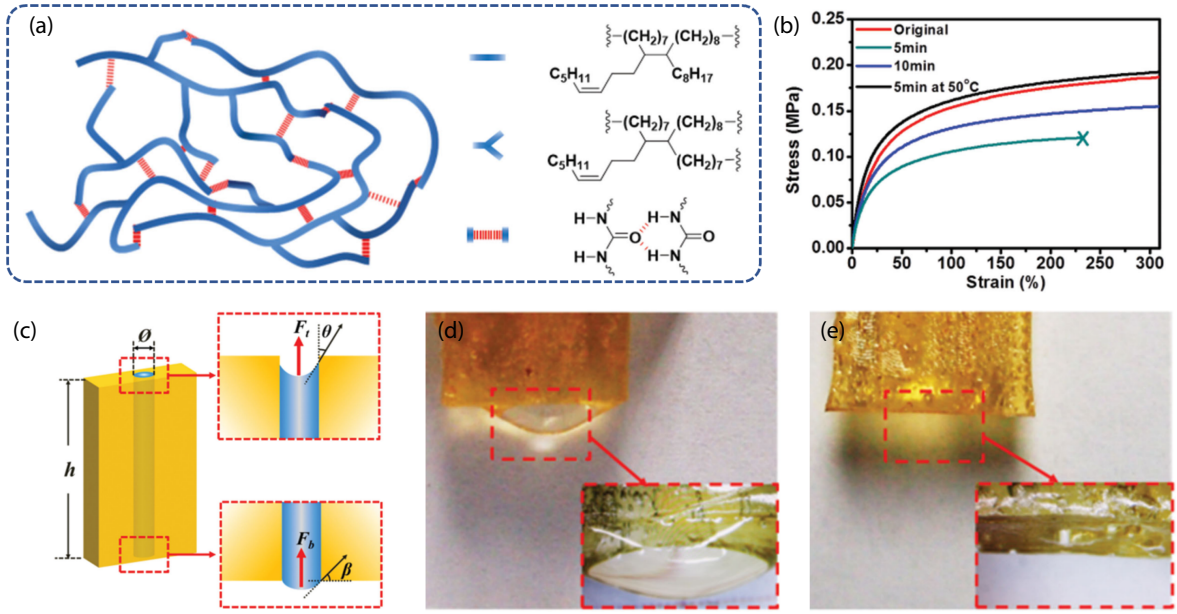


Fig. 1 (a) Schematic illustration of SHPs based on hydrogen bonding interaction; (b) Stress-strain curves of original SHP sample and broken samples healed for 5 and 10 min at room temperature or 5 min at 50 °C; (c) Schematic illustration of ionic liquid trapped in a vertical microchannel *via* capillary force; (d, e) Photographs of [OMIm][PF_6] enclosed in the microchannels with diameter of (d) 1.615 mm and (e) 0.709 mm. (Reproduced with permission from Ref. [62]; Copyright (2015) WILEY-VCH Verlag GmbH & Co. KGaA.)

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trolling appropriate channel size based on theoretical modeling. When a SHP loaded with a small column of ionic liquid was broken from the middle part, a new port was opened while the ionic liquid would not leak out due to the barometric pressure. However, unexpected damage of the device may lead to worse cases that both ends are opened. In order to avoid leakage of ionic liquid in such an extreme condition, a series of theoretical experiments have been designed, wherein the SHP microchannel filled with ionic liquid was vertically placed and the top and bottom ports were both opened. As shown in Fig. 1(c), the ionic liquid tended to flow downwards due to the gravity effect, which could only be overcome by the capillary force. Hence, the maximum capillary force that could be provided by the microchannel decided the possibility of leakage during the unexpected fracture or damage. At this point, there are two major interfaces that provide capillary forces, including the top end (F_t) and bottom end (F_b). According to Eqs. (1) and (2), the maximum capillary force provided by the microchannel as well as the maximum height of liquid column can be calculated.

$$F_{c.max} = \pi\Phi \cdot \gamma_{g,l}(\cos\theta + \sin\theta) \quad (1)$$

$$h_{max} = \frac{4\gamma_{g,l}(\cos\theta + \sin\theta)}{\rho g \Phi} = \frac{16.11}{\Phi\,(\text{mm})}\,(\text{mm}) \quad (2)$$

According to above equations, the maximum capillary force that the microchannel could provide is determined by the diameter of the microchannel (Φ), surface tension of the ionic liquid ($\gamma_{g,l}$), and contact angle between the ionic liquid and the SHP matrix. Thus, referring to a specific ionic liquid (1-methyl-3-octylimidazolium hexafluorophosphate, [OMIm][PF_6]), the maximum height of liquid column that the microchannel could hold is only dependent on the diameter of this channel, suggesting that the possibility of leakage could be theoretically controlled by minimizing the channel size. This equation has also been verified *via* related experiments. In detail, [OMIm][PF_6] was injected into two microchannels with diameters of 1.615 and 0.709 mm, respectively, and the liquid column length was fixed at 15 mm. When the SHP matrix was vertically placed, [OMIm][PF_6] flowed out of the wider channel while still retained in the narrower channel (Figs. 1d and 1e).

Inspired by liquid confinement effect in the microchannels, two major types of self-healing ionic liquid-based electronic devices have been fabricated with the assistance of the SHP matrix. Based on our previous discoveries on ionic liquid, two principles of temperature-dependent electrical behaviors of ionic liquids were applied for building electronic devices in response to temperature variations. On one hand, the conductivity of the ionic liquid dramatically increases with tiny temperature increment, suggesting that it can serve as an ultra-sensitive thermometer or as the media to detect other forms of energy, like light or alternative magnetic field. On the other hand, the absorption behavior of ionic liquid on the surface of electrode varies as the temperature changes, leading to unusual thermoelectric behavior that can be applied as thermometer or exploited for energy transfer and storage. In the following sections, we would like to thoroughly introduce the basic design and performance of the two types of self-healing ionic liquid-based electronic devices.

Owing to the outstanding thermal sensitivity of ionic liquids, the SHP matrix with a tubular channel loading [OMIm][PF_6] was exploited as a self-healing electronic thermometer.[62] As shown in Fig. 2(a), within a body temperature range of 35–45 °C, the [OMIm][PF_6]-based thermometer exhibited excellent sensitivity, showing a 7% signal change upon temperature variation of only one degree. Notably, in combination of seal-healing ability of the SHP matrix and the ionic liquid, the thermometer exhibited electrically healing as well as mechanically repairing abilities. Typically, the continuous conductive circuit of ionic liquid was cut off along with the broken of the outer SHP matrix. Once the two broken parts were rejointed, the resistance of the ionic liquid circuit could instantly recover to its original level (Fig. 2b). Similarly, even if the thermometer encountered breaking during the sensing tests, it could also recover to the original state and continue to monitor the temperature signal as before (Fig. 2c). Except for thermal sensing applications, owing to the ease of doping in the liquid materials, other functional sensing elements could be easily doped into the ionic liquids, broadening more functional sensing applications of this self-healing ionic liquid-based electronic devices. For example, attributing to the strong cation-π interaction, carbon nanotubes, as a kind of dark material with high photothermal conversion efficiency, could be well dispersed into ionic liquids, serving as a media to transfer the light signal to temperature change. Thus, the CNT-doped ionic liquid was successfully applied as a photodetector with ultrahigh sensitivity. By integrating the CNT-doped [OMIm][PF_6] into the SHP matrix with a serpentine microchannel (Fig. 2d), a self-healing near-infrared (NIR) photodetector was fabricated that showed a direct and ratiometric response to the NIR light with varied power densities. Following the same self-healing principle as sated above, this NIR photodetector also recovered its function and sensitivity after its rejointing from the broken parts (Fig. 2e). In addition, such a strategy could also be extended to detect the radio wave *via* doping the iron(II, III) oxide nanoparticles (Fe_3O_4) into the ionic liquid, and further exploited as a self-healing radio sensor. The conductivity of the ionic liquid would increase along with the heat generation by the magnetothermal conversion of these paramagnetic nanoparticles.

Besides conductivity change as the temperature varies, the adsorption of ionic liquid on the surface of electrode is strongly dependent on the environmental temperature. By asymmetrically heating or cooling two electrodes in an ionic liquid circuit, we presented the thermoelectric effect of ionic liquid several years ago.[54,63] Typically, the ionic liquid was injected into a capillary tube and sealed with two gold wires as electrodes. After heating one side of the circuit, the circuit could output a Seebeck voltage with a highest Seebeck coefficient of 1.446 mV/K for 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide ([EMIm][Tf_2N]), which is much higher than those of most semiconductors. Specifically, the [EMIm][Tf_2N] output a voltage over 60 mV under a temperature difference about 44 K. In combination with SHP matrix, a self-healing thermoelectric device was fabricated, presenting

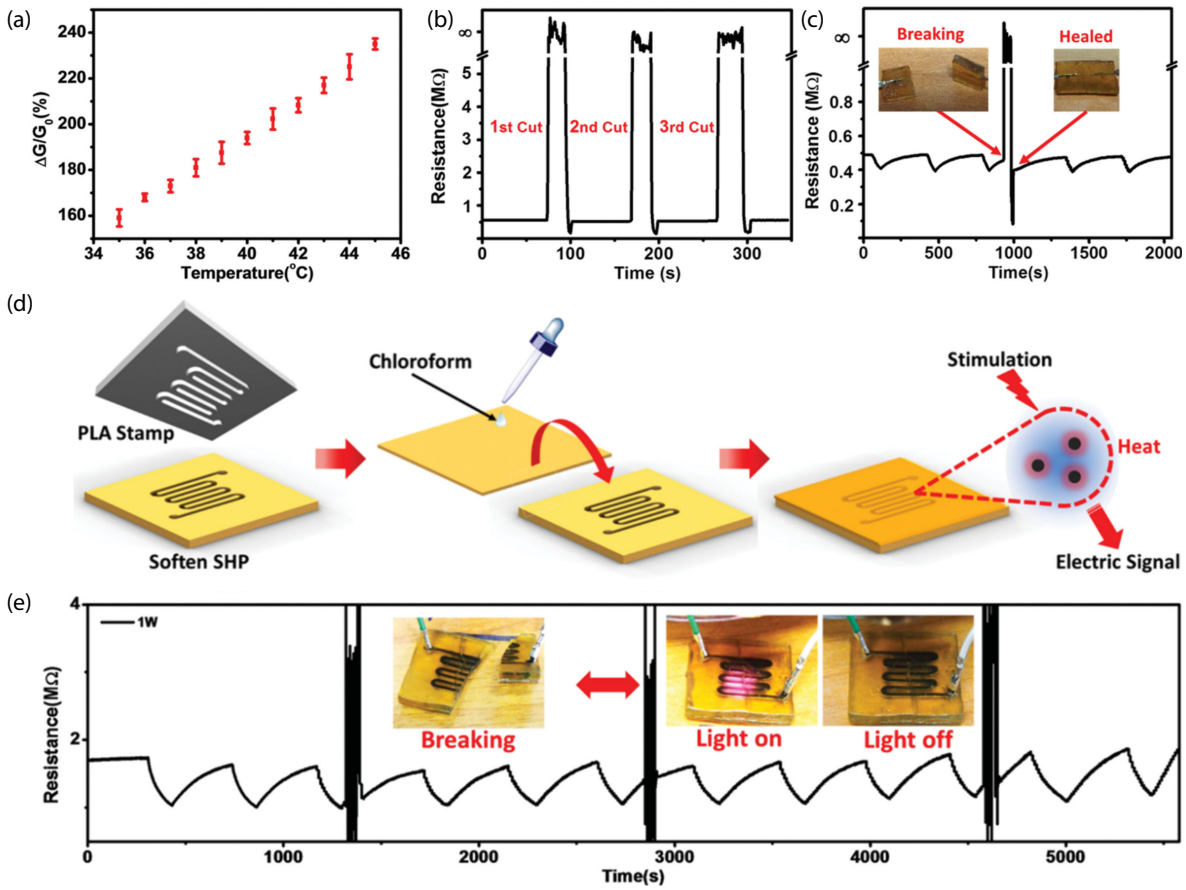


Fig. 2 (a) Thermal response of [OMIm][PF_6] loaded in the channel of SHP matrix at temperature ranging from 35 °C to 45 °C. (b) Resistance changes during several cutting-healing cycles; (c) Resistance changes over several heating-cooling cycles, during which the sensor was cut off and then rejointed for healing; (d) Schematic illustration of the fabrication procedure for near infrared sensors based on a serpentine microchannel and carbon nanotube-doped [OMIm][PF_6]; (e) Resistance changes over several NIR light on-off cycles, during which the sensor was cut off and then healed for three times. (Reproduced with permission from Ref. [62]; Copyright (2015) WILEY-VCH Verlag GmbH & Co. KGaA.)

a similar Seebeck voltage as the capillary chip (Fig. 3a). During the cyclic tests, if the ionic liquid-based thermoelectric device was cut off, the signal of output voltage became unable to be detected. Once two broken parts were reunited, the output voltage immediately recovered and exhibited no difference during cyclic heat on-off tests in comparison with that before broken, indicating effective healing ability of the thermoelectric performance (Figs. 3b–3d). This profound breakthrough has never been observed in other semiconductor-based thermoelectric systems.

INTRINSICALLY CONDUCTIVE, SELF-HEALING, AND RECYCLABLE IONIC LIQUID-BASED ELECTRONICS

Although electronic devices made of ionic liquid enclosed in self-healable polymer microchannels present outstanding self-healing ability, it is unable to realize full recovery once it encounters fatal damage and completely breaks apart. Due to the spatial separation of ionic liquid and SHP matrix, the liquid is unavoidable to leak out from the channel while the whole matrix was broken apart. Thus, new strategies that homogenize ionic liquids with self-healing polymer matrix are anticipated to efficiently avoid the liquid leakage and enhance the self-healing ability, or even the recyclability. In this regard, two major approaches have been demonstrated in our recent work, including chemical grafting of ionic liquid onto the self-healing polymer chains and physical confinement of ionic liquid in crystal microplates. Both strategies have been applied for synthesizing a series of intrinsically conductive ionic liquid-based materials with superior self-healing ability and recyclability, which are capable of being produced into electronic devices without the assistance of external encapsulation materials.

According to the idea of chemical grafting, a kind of intrinsically recyclable ionic liquid-based electronic material (poly(UHH-*co*-IL)) was synthesized.[64] In detail, 1-vinyl-3-ethyl-imidazolium acetate ([VEIm][Ac]) was copolymerized with another vinyl monomer (UHH) containing the quadruple hydrogen bonding motif (2-ureido-4-[1*H*]pyrimidinone, UPy), forming a conductive network crosslinked by the quadruple hydrogen bonds (Fig. 4a). Interestingly, [VEIm][Ac] exhibited a relatively high solubility to UHH, and thus became the ideal candidate for serving as both monomer and solvent of UHH during polymerization. With the increasing amount of UHH as

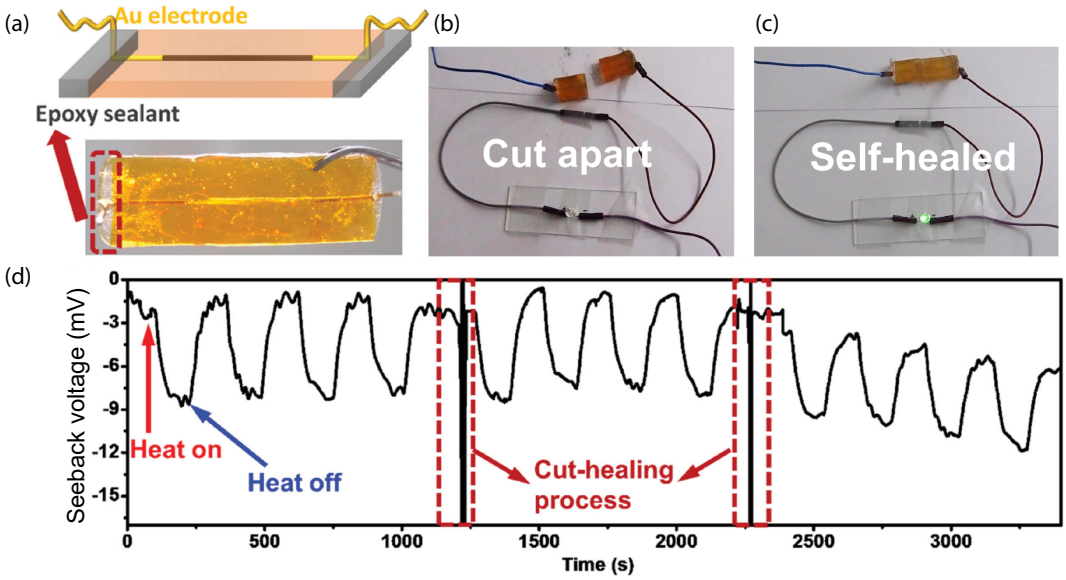


Fig. 3 (a) Schematic illustration and photograph of the self-healing [EMIm][TF_2N]-based thermoelectric device. (b, c) Photographs of the self-healing thermoelectric device connected with a LED: (b) broken state, (c) healed state. (d) Seeback voltage over cyclic heat on and off, during which the device was cut off and healed twice. (Reproduced with permission from Ref. [63]; Copyright (2016) WILEY-VCH Verlag GmbH & Co. KGaA.)

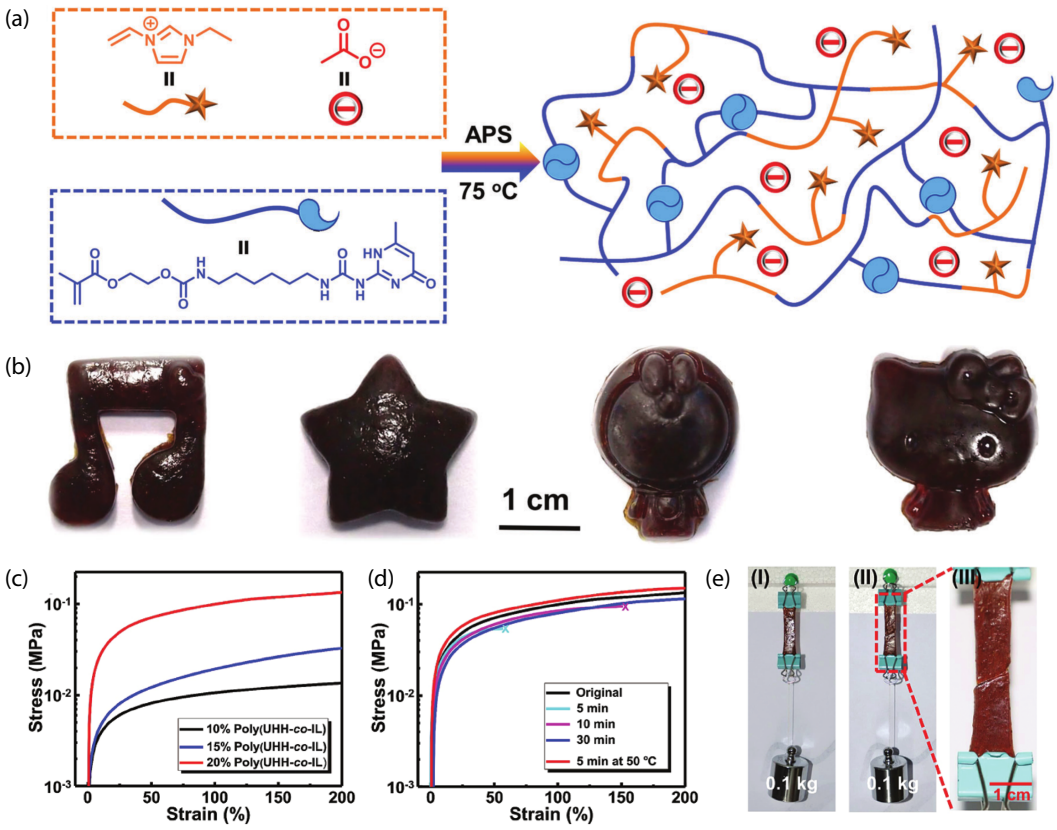


Fig. 4 (a) Schematic illustration of the fabrication process of poly(UHH-*co*-IL). (b) Photographs of poly(UHH-*co*-IL) samples in different shapes thermally processed by different molds. (c) Stress-strain curves of poly(UHH-*co*-IL) with UHH molar ratio of 10%, 15%, and 20%. (d) Stress-strain curves of original sample of poly(UHH-*co*-IL) with UHH molar ratio of 20% and broken samples healed for 5, 10, and 30 min at room temperature or 5 min at 50 °C, respectively. (e) Photographs of poly(UHH-*co*-IL) film holding 0.1 kg weight: (I) original sample; (II) broken sample healed at room temperature for 30 min; (III) enlarged image of (II). (Reproduced with permission from Ref. [64]; Copyright (2018) The Royal Society of Chemistry.)

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well as hydrogen bonds, mechanical strength of poly(UHH-*co*-IL) was reinforced (Fig. 4c). Owing to the reversibility of hydrogen bonds at high temperatures, this copolymer could be easily reprocessed *via* thermal molding procedure. For example, as shown in Fig. 4(b), poly(UHH-*co*-IL) was molded into various shapes, verifying its excellent thermal processing ability and recyclability. With the strategy of copolymerization, the conductivity and thermal sensitivity of the ionic liquid could be maintained as well as the mechanically healing and recycling ability, suggesting that poly(UHH-*co*-IL) might fully meet the requirement for fabricating intrinsically self-healing and recyclable ionic liquid-based electronic sensors. Typically, mechanical performance was fully recovered after being rejointed for only 30 min at room temperature or within 5 min at 50 °C (Figs. 4d and 4e). On the other hand, the electrical property instantly recovered to that of original state once the two broken parts were brought into contact. Notably, both resistance and thermal sensing tests exhibited good repeatability during several consecutive breaking-healing cycles (Figs. 5a–5c). All the above results prove the excellent self-healing ability of poly(UHH-*co*-IL) in mechanical, electrical, and thermal sensing aspects, which fully meets the requirements for an intrinsically self-healing thermal sensor.

Beyond self-healing, the quadruple hydrogen bond that is sensitive to temperature facilitates the thermal reprocessing or recycling of poly(UHH-*co*-IL), suggesting that the electronic sensors made of poly(UHH-*co*-IL) could be recycled even after encountering a catastrophic damage (Fig. 6a). A forehead temperature thermometer made of poly(UHH-*co*-IL) was grinded and remolded for 4 times, during which the temperature monitoring results were fully consistent with the value measured by an infrared thermal camera (Fig. 6b). In addition, the excellent reprocessability of poly(UHH-*co*-IL) could be utilized for fabricating customized electronic devices. As a practical demonstration, customized ear canal thermometer based on poly(UHH-*co*-IL) was fabricated *via* multiple molding process and applied for monitoring the ear canal temperature (Figs. 6c and 6d). Similarly, the customized ear canal thermometer could also be recycled and remolded into a new one that fitted the ear canal of another person. As shown in Fig. 6(e), both the original and recycled ear canal thermometer provided a temperature value precisely close to the results captured by the infrared thermal camara. In a short summary, the chemical strategy of copolymerizing ionic liquid with monomer containing quadruple hydrogen bonds paves a new way in designing self-healing and recyclable electronic materials and devices. Except for copolymerization, ionic liquids could serve as the dopant in self-healing polymer network, constructing intrinsically conductive and self-healing ionogel materials.[67–69] For example, Sun and coworkers[67] reported a series of ultra-durable ionic skins with excellent healability and sensitivity based on a self-healable polyurethane dissolved with 1,2-dimethyl-3-ethoxyethyl-imidazolium bis(trifluoromethanesulfonyl)imide ([DEIM][TFSI]). Taking the hindered urea bond as the self-healing moiety, the fractured ionic skins could be easily repaired at 65 °C and restore their original ultra-durable sensing performance. It is notable that the design of this ultra-durable ionic skin can be extended to fabricate a wide variety of self-healable and highly sensitive electronic sensors.

From the viewpoint of self-healing capability in electronics, the intelligent liquid robot named T-1000 in *Terminator 2: Judgement Day* has been widely recognized as the ideological and ultimate style of electronic devices, which can reshape

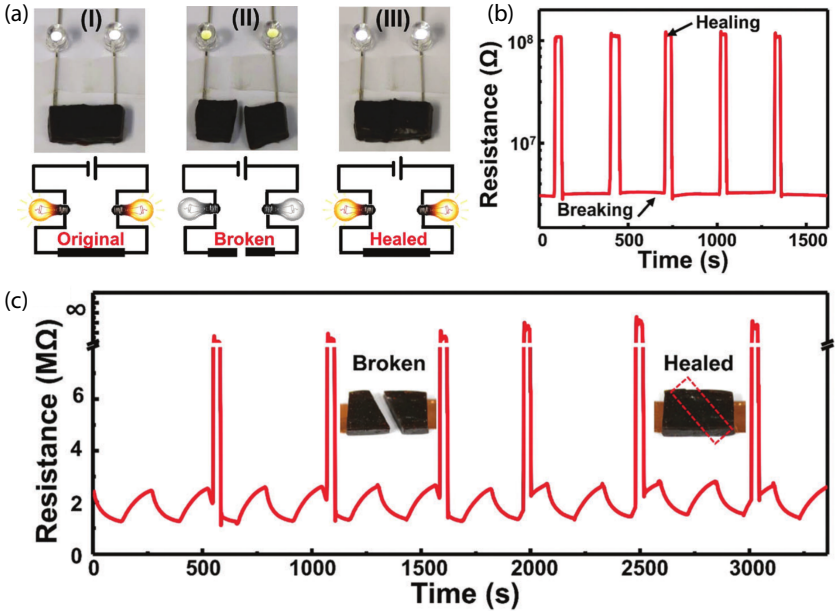


Fig. 5 (a) Photographs and schemes of poly(UHH-*co*-IL) connected in series with two white LED indicators: (I) original state; (II) broken state; (III) healed state. (b) Resistance change of poly(UHH-*co*-IL) during several cutting-healing cycles. (c) Resistance change of poly(UHH-*co*-IL) thermal sensor over several heat on-off cycles, during which the sensor was cut off and then healed for several times. (Reproduced with permission from Ref. [64]; Copyright (2018) The Royal Society of Chemistry.)

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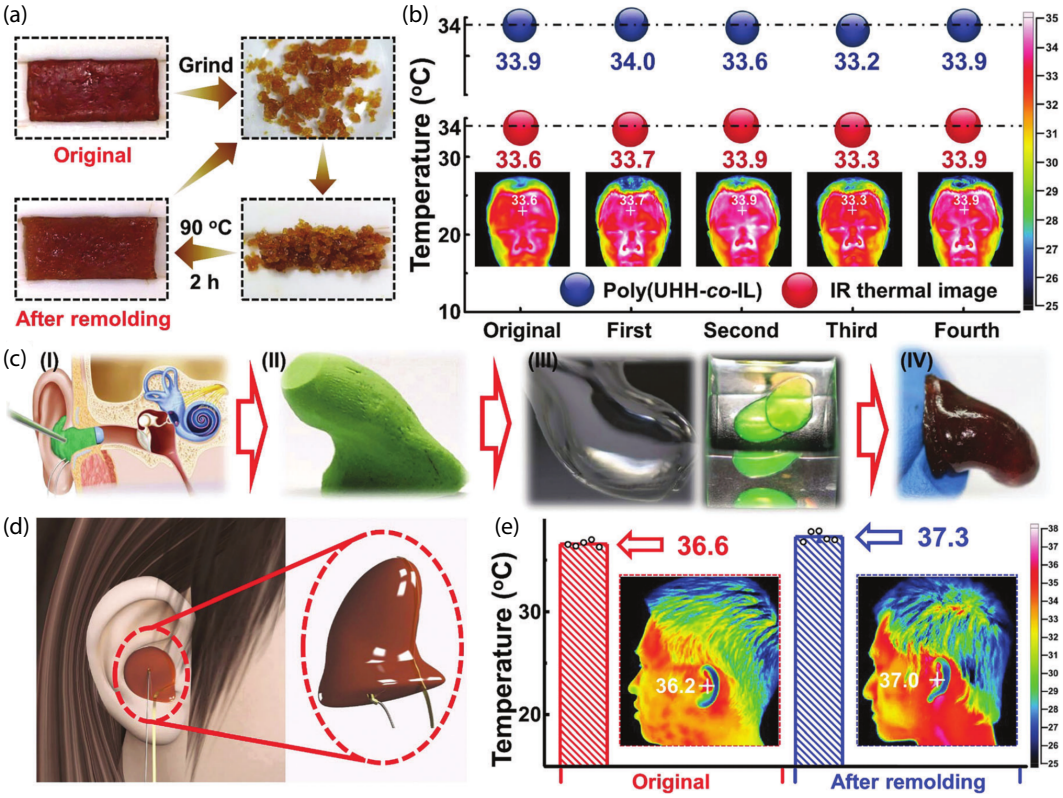


Fig. 6 (a) Photographs of poly(UHH-*co*-IL) samples at different states in a recycling process. (b) Forehead temperature measured by poly(UHH-*co*-IL) thermal sensor recycled for different times and IR thermal camera. Insets are thermal images captured by an IR thermal camera. (c) Scheme and photographs demonstrating the process of fabricating the customized thermal sensing earplug. (d) Schematic illustration of monitoring ear canal temperature *via* the customized earplug sensor. (e) Ear temperature of two volunteers measured by the original and re-customized earplug sensor. Insets are thermal images captured by an IR thermal camera. (Reproduced with permission from Ref. [64]; Copyright (2018) The Royal Society of Chemistry.)

to pretend as different people and rapidly repair once encountering damage. Aiming to exploit ionic liquid-based electronic devices like T-1000, our group proposed a physical strategy based on the crystal confinement effect.[65] Similar to the chemical strategy stated above, the crystal confinement strategy could not only maintain the thermal sensitivity of ionic liquid, but also avoid the risk of leakage even when it encounters damage. Typically, a flowing ionic liquid [OMIm][PF_6] was mixed with a kind of ionic liquid crystalline containing azobenzene group ([OMIm][AzoO]) at its melting temperature around 70 °C. Due to the strong π-π stacking between the azobenzene group, [OMIm][AzoO] tended to precipitate and crystalize once the temperature drops below its melting point (Fig. 7b), forming the crystal microplates that could effectively confine the [OMIm][PF_6], which was termed crystal-confined ionic liquids (CCILs). According to the prediction based on the capillary confinement effect, the theoretical confinement height in a glass tube (3 mm in diameter) exceeded one meter when the weight ratio of [OMIm][AzoO] reached 60%, indicating a much better sealing effect for flowing ionic liquid in comparison with the previous microchannel approach. Similarly, as the increasing amount of [OMIm][AzoO], thermal sensitivity of the CCILs composite increased, which reached 890% for the composite with 60 wt% [OMIm][AzoO] when the temperature ramped from room temperature to 60 °C (Fig. 7b). Notably, this conductivity change is the highest value among all kinds of organic thermal sensing materials as far as we know.

Unlike polymeric material, the melting of small molecular aggregates was much quicker, and the binary ionic liquid composites turned into fluid once heating over the melting point, thus realizing efficient self-healing, molding, or even complete reconfiguration (Fig. 8a). For example, under stimulation of heating, a human-shape conductor made of CCILs could quickly shift from one circuit into another one and subsequently trigger the function of the new circuit (Fig. 8b). Moreover, owing to the excellent fluidity in the melted state, the electronic devices made of CCILs could also repair itself like the T-1000 after encountering damage. As shown in Fig. 8(c), in a practical simulation, an artificial robot arm made of CCILs was fixed on a wooden figure. When the arm encountered external damage, the resistance of the circuit would increase and then trigger local heating by a thermal heater. Once the resistance recovered to a threshold that suggests the complete healing, the heating circuit was automatically cut off. As shown in Fig. 8(d), the CCILs arm successfully healed itself without manual intervention, just like the Terminator robot, which could greatly push the forefront of self-healing electronics to reach the goal of this science-fiction movie.

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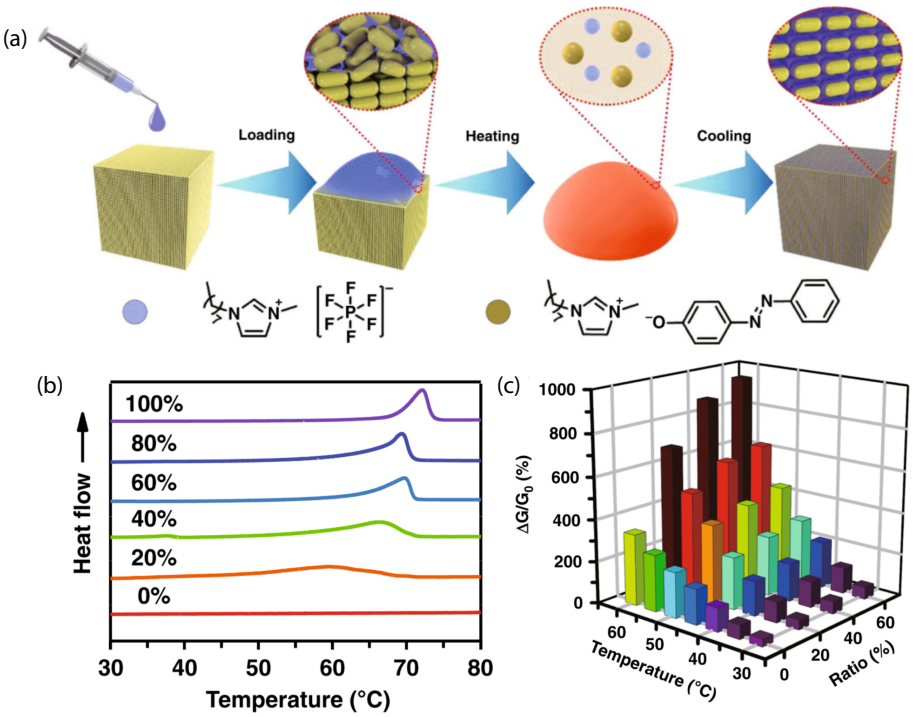


Fig. 7 (a) Schematic illustration of preparing crystal-confined ionic liquids (CCILs). (b, c) Differential scanning calorimeter (DSC) curves and thermal sensing performance of CCILs with different weight percentages of [OMIm][AzoO]. (Reproduced with permission from Ref. [65]; Copyright (2019) Springer Nature.)

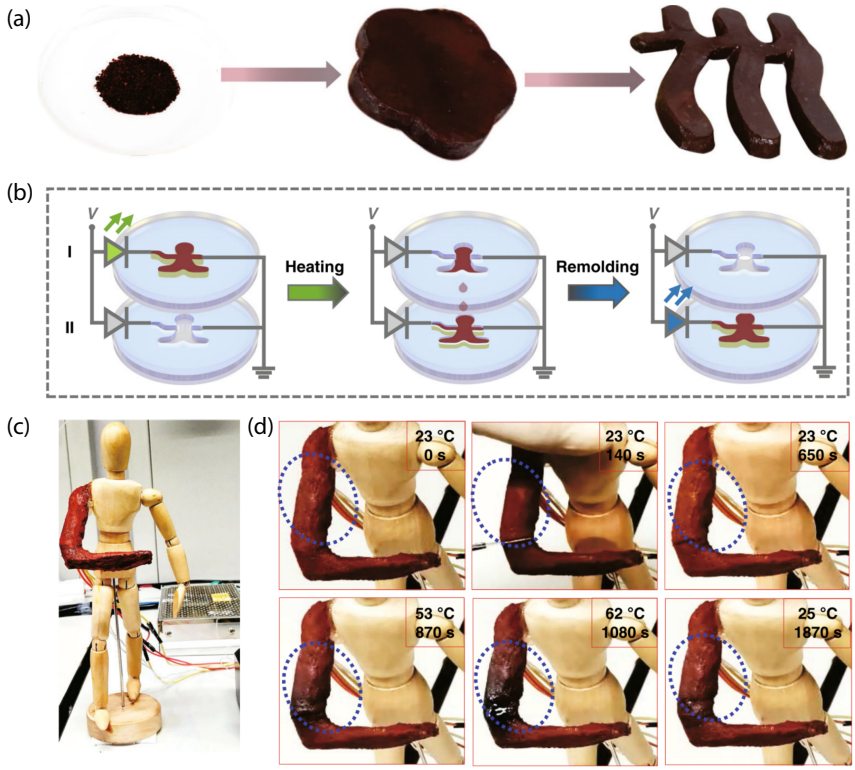


Fig. 8 (a) Freestanding CCILs (60 wt% of [OMIm][AzoO]) with different shapes, from left to right: powderlike product, plum blossom, and simplified logo of the Renmin University of China. (b) Schematic illustration of transporting CCILs containing 60 wt% of [OMIm]AzoO from the upper circuit to the bottom circuit. (c) Photograph of a wooden figure patched with CCILs, which connects with a temperature controlling system. (d) Photographs of different status during the cutting and healing process, including without injury (0 s), suffering cutting (140 s), and healing (140~1870 s). (Reproduced with permission from Ref. [65]; Copyright (2019) Springer Nature.)

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CONCLUSIONS AND PERSPECTIVES

In this feature article, we review our recent progresses in the aspect of self-healing electronics based on ionic liquids which offer fascinating promise for prolonging the service life of electronic devices as well as reducing electronic waste. Focused on the viewpoint of avoiding the leakage problem of ionic liquid, two comprehensive approaches have been particularly emphasized. One is to enclose ionic liquids in the microchannel made of self-healing polymers. The strong capillary effect accounts for ionic liquids held by the microchannels at opened states. With this strategy, ionic liquid-based thermometers and thermoelectric devices were fabricated, which exhibited prominent self-healing performance without the sacrifice of the sensing or thermoelectric performance. The microchannel-confinement approach provides a versatile method for fabricating various kinds of functional self-healing devices with ionic liquid or even other liquid electrolytes as conductive or sensing elements. However, it is notable that the self-healing ability of this system is relatively limited due to the unavoidable leakage during catastrophic damage, and the polymer matrix-assisted devices cannot be easily reprocessed or recycled into a new one. In comparison, another approach is to homogeneously disperse ionic liquids in a chemical or physical framework, such as copolymerizing into a polymer network or being confined in another ionic liquid crystals. Taking the advantages of homogeneity, this type of electronic devices exhibited excellent self-healing and recycling ability without any loss of the electrical sensing performance. However, this approach requires sophisticated chemical or material structure design and synthesis, hardly to be facilely applied for different liquid sensing elements.

In spite of great progresses that have been achieved in the last few years, further improvement of ionic liquid-based electronics is still intertwined with plenty of challenges and remaining issues. Thus, in the last part of this article, we would like to address these challenges or issues and share our perspectives of self-healing ionic liquid-based electronics from the aspects of fabrication and performance. (1) The fabrication procedure of both self-healing materials and ionic liquid-based electronic devices are incompatible with current electronic technologies, like photolithography, micropatterning, and printing technique. Developing self-healing materials and ionic liquid circuits that are applicable to these techniques can effectively improve the degree of integration for ionic liquid-based electronic devices. With improved integrity, ionic liquid electronics may be exploited in more functional applications, such as integrated circuits. (2) The resistance of the ionic liquid-based devices is much higher than that of traditional conductors or even part of the semiconductors, which generally requires complicated measurement devices and thus severely restrict its popularization. More efforts should be contributed to synthesizing new types of ionic liquids with high ionic mobility or doping compatibility for enhancing the conductivity. (3) As most self-healing and recyclable materials are related with thermally-triggered non-covalent bonds, the thermal stability of the self-healing ionic liquid-based electronic devices should be taken into account. Ideally, the working temperature range of the electronic devices should be completely separated with the healing or recycling temperature range, achieving an orthogonal condition between working and recycling, and improving the thermal stability of the electronic devices. In addition, photo-triggered repairing or recycling components are suggested to be incorporated, which can provide more convenient manipulation methods for smart ionic liquid-based electronics. In future, as the development of ionic liquid-based self-healing materials and improvement of integrity of ionic liquid circuits, we may finally fulfill the functions of liquid robot T-1000 with ionic liquid-based electronics. It is for sure that ionic liquid-based electronics would play an irreplaceable role in soft electronics and share a piece of cake in the electronic industry.

BIOGRAPHY

Yapei Wang is a full professor at the Department of Chemistry, Renmin University of China. He received his B.S. from Jilin University in 2004 and Ph.D. from Tsinghua University under the supervision of Prof. Xi Zhang in 2009, both in chemistry. After graduation, he spent 2 years as a postdoctoral fellow in Prof. Joseph M. DeSimone's laboratory at the University of North Carolina at Chapel Hill. In 2012, he joined the Department of Chemistry, Renmin University of China. His research interests are mainly focused on photothermal conversion materials, liquid electronics, and interface science.

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