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REVIEW ARTICLE

An Overview of Material Solutions to Solve Self-Healing and Economic Challenges in the Development of Electronic Skin

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ABSTRACT: Human skin allows us to experience sensations by converting physical contact and movement into interpretable electrical impulses for the nervous system to read through specialized neurons called mechanoreceptors. Electronic skin (e-skin) attempts to emulate this process by creating a network of flexible materials that generate electrical feedback like mechanoreceptors. E-skin material choice is diverse, as each material has unique properties that the e-skin inherits. Thus, material choice is paramount in developing e-skin to solve challenges. As a result, this review explores challenges in e-skin development and material innovations to solve challenges with self-healing and sustainable e-skin development. This review will analyze and compare different materials used in recent research to determine the kinds of materials for researchers to prioritize in future research. Developing commercially sustainable e-skin with self-healing ability will lead to breakthroughs in medicine, intelligent robotics, augmented reality, and more.

KEYWORDS: Biomedical Engineering, Biomedical Devices, Electronic Skin; Sustainable Design, Self-Healing.

Introduction

The development of commercially sustainable electronic skin (e-skin) has spurred innovative material solutions to e-skin development challenges. E-skin refers to flexible electronic devices capable of one or more sensing functions, made to achieve high-sensitivity detection of its external environment. E-skin offers superior flexibility and thinness than traditional sensors, which increases their versatility.[1] These novel materials have applications in intelligent robotics, medicine, augmented reality, and more.[2-4]

Typical e-skin designs combine flexible materials that can sense along a single path.[1] E-skin sensing capabilities must function correctly and produce consistent results despite mechanical duress such as stretching and bending, which require flexible or soft sensing material.[5] In the past, e-skins utilized sensor arrays to detect tactility and pressure.[6] However, sensor arrays are often limiting factors on the size of e-skins, as they can quickly become complex.[6] Their sensing resolution is also limited by the number of cells in the array.[6]

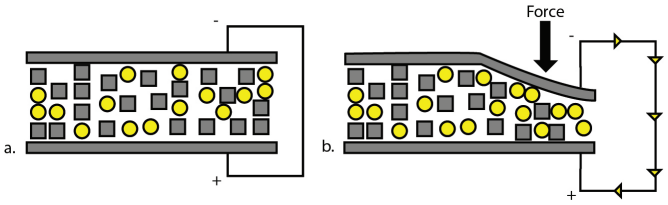

Figure 1a and 1b: Two diagrams detailing a piezoresistive material and the alignment of ions within their respective molecular structures. A force on the material, typically a mechanical force, changes the alignment of ions within the material's structure, temporarily changing the electrical resistance of the material and allowing a current to be formed.

Research has progressed to other methods of sensing through materials, piezoresistive in particular, but also including piezoelectric, and triboelectric materials, which change resistance or current in response to physical stimuli as seen in Figure 1a and 1b.[7] Software interprets the change in resistance or current, resulting in signal detection.[7] Piezoresistive materials are simple to construct, have low-power consumption, and simple readout mechanism.[8]

The material chosen for the e-skin determines its physical properties. E-skin research has led to the development of new materials as the challenge e-skin presents is unique; the material must be lightweight, flexible, elastic, nontoxic, and thermally and chemically stable.[2,9] E-skins also face challenges including their ability to self-heal and economic viability.[10]

This review will highlight material-based solutions to the challenges mentioned above – other currently existing challenges are beyond the scope of this paper.[10] This review also aims to create a resource for new researchers to learn about current material innovations in the field, analyze each material's strengths and weaknesses, and identify where they should focus their efforts. The continued progress of e-skin, combined with the advancement of AI technologies, will open opportunities for new advancements in intelligent robotics.[11]

Additionally, this paper argues that self-healing and cost-effective e-skins are critical challenges to the future of e-skin development and merit continued research. This paper asserts that further research should be conducted into crosslinking self-healing mechanisms that can operate underwater and into searching and testing naturally abundant and sustainable polymers like chitin for sustainable e-skin development.

The review will introduce the ability of e-skin to self-heal and highlight current developments in self-healing hydrogels and elastomers. Self-healing hydrogels and elastomers were chosen over other self-healing material options because of their versatility and ability to fulfill the aforementioned goals. Finally, the review will introduce recent solutions to creating sustainable and economically viable e-skins for commercial use.

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Discussion

1. Self-Healing:

Damage to e-skins from intense mechanical stress is inevitable, leading to decreased performance, failure, and costly maintenance.[12-14] Self-powered e-skins are typically multi-layered, consisting of a heterogeneous triboelectric layer and electrode layer that lack mechanical synergy due to differing material properties.[12] Thus, they are prone to mechanical damage and require self-healing to be sustainable over long periods of use.[14,15] Self-healing materials offer a solution to this challenge. Adapting self-healing properties for e-skin is difficult as it often requires two materials with different properties to work together to self-heal.[15] Recent self-healing e-skins have faced issues with thickness, which decreases sensitivity and flexibility – two key characteristics of a fully functional e-skin.[17] Two kinds of materials, hydrogels, and elastomers, have emerged as solutions to these challenges because of their excellent mechanical and electrical properties, self-healing potential, and biocompatibility.[17]

1.1. Elastomers:

Elastomers are a popular material in self-healing development for e-skin. Elastomers are polymers characterized by elastic properties made of crosslinked chainlike molecules.[18] This allows them to elongate and stretch under duress without losing shape.[18] Their mechanical strength, flexibility, and stretchiness make them suitable candidates for e-skins.[14] However, many elastomers cannot repair themselves after significant damage, so researchers have begun investigating methods to create self-healing elastomers.

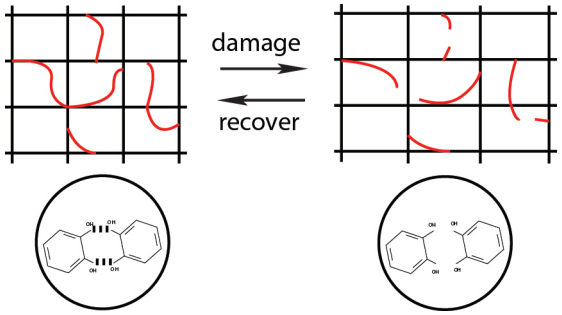

Figure 2: A diagram showing the mechanism by which hydrogen-based self-healing methods relink after undergoing damage. After undergoing mechanical damage, dynamic bonds reform to heal the material.

Creating self-healing elastomers that combine both mechanical strength and self-healing efficiency requires a self-healing mechanism.[19] A recent review by Utrera-Barrios *et al.* categorized recent self-healing elastomers into three generations: elastomers requiring a healing agent that releases upon damage to begin self-healing, elastomers relying on dynamic bonds, and elastomers utilizing vascular networks. Vascular network-based elastomers have not found much success because of difficulties incorporating them into the elastomer.[19] Elastomers relying on dynamic bonds have received much attention because of their ability to self-heal without extrinsic mechanisms (Figure 2).

1.1.1. Materials:

Griggs *et al.* chose thermoplastic polyurethanes (TPU) to create a self-healing e-skin.[20] TPUs were chosen for their versatility and self-healing properties.[20] The self-healing mechanism was based on hydrogen bonding, disulfide bonding, and van der Waals forces to induce self-healing at ambient temperatures without outside stimulus.[20] They created several TPUs and found that P40 exhibited the strongest self-healing qualities.[20]

Xun *et al.* used conductive and insulated polyurethanes (PUE) to create an e-skin with self-healing capabilities and triboelectric properties.[12] Triboelectric e-skins often consist of a triboelectric layer and an electrode layer, making self-healing difficult in the two materials.[12] Their goal was to create a self-healing e-skin that could heal two layers of materials.[12] They combined insulated PUE acting as the triboelectric layer and conductive PUE acting as the electrode layer. They reasoned that the homogeneous structures and similar properties of conductive PUE and insulated PUE would allow for self-healing and stability.[12]

Liu *et al.* also created a triboelectric self-healing e-skin using polytetramethylene ether glycol (PTMEG).[16] They chose PTMEG for its ultra-thinness, transparency, and exhibition of nanoscale phase separation.[16]

However, none of the listed e-skins reported the ability to function in wet environments.[12,14,20] This is most likely because their self-healing mechanisms were based on polar interactions that would be interrupted by polar fluid.[12,14,20] In an attempted solution, Kang *et al.* reported a water-insensitive self-healing elastomer based on polydimethylsiloxane (PDMS).[14] Interestingly, the self-healing mechanism was based on hydrogen bonds. Kang *et al.* hypothesized that the structure of PDMS made hydrogen bonds energetically favorable when immersed in water.[14] A separate study by Kong *et al.* engineered a underwater self-healing material using this idea.[21] They strategically designed their elastomer to be unaffected by submerged conditions.[21]

1.1.2. Results and Discussion:

After 20 minutes, Griggs *et al.*'s TPU P40 showed 69±2% self-healing efficiency of elongation and 101±3% of tensile strength. After 3 hours, it rose to 91±4% and 108±14% respectively.[20]

After 24 hours, Xun *et al.*'s e-skin fully self-healed a 50 nm wide cut.[12] They found that the dynamic bonds had undergone reconstruction across the damaged surface.[11] After 24 hours at 25 °C, they measured 71% restoration of elasticity and 92% restoration of mechanosensing capabilities.[12] The PUEs self-healing efficiencies were around 20% lower than the TPUs. However, they found the self-healing efficiency increased when an external heat source was applied. After 24 hours at 75 °C, the elasticity and mechanosensing restoration increased to 92% and 97%, respectively.[12]

After 24 hours at 50 °C, Liu *et al.* found their e-skin could reach 90% restored mechanical strength.[16] Similar to the PUE e-skin, the PTMEG e-skin retained its self-powering properties after self-healing.[16]

Kang *et al.* produced an e-skin that self-healed even when immersed in water or artificial sweat. After 48 hours, they reported a 78% self-healing efficiency, notably lower than the self-healing efficiencies of the elastomers mentioned above.[14] They were still determining why PDMS could self-heal in water.[14] They hypothesized that the hydrophobicity of the PDMS backbone increased the enthalpy gain for hydrogen bonding formation, making it energetically favorable for the hydrogen bonds to reform.[14] They also reported insignificant water uptake, allowing for electronics to be embedded in the PDMS e-skin without fear of water damage when submerged.[14]

The TPU reported the highest self-healing efficiency in the least time without external energy, outclassing the other elastomers by large margins. Thus, for pure self-healing efficiency, TPUs are an optimal material for implementing self-healing into e-skins. Despite Kang *et al.*'s subpar self-healing efficiency at 78%, its ability to self-heal while submerged in liquid is significant. It is foreseeable that e-skin will be used in fields where exposure to water is inevitable. As a result, it is worth the effort to investigate why PDMS can self-heal in liquid; concrete research will provide key information that will allow researchers to identify other materials that can self-heal when submerged. Further understanding could also lead to PDMS's improved self-healing efficiency.

1.2. Hydrogels:

Hydrogels are a class of soft materials characterized by porous hydrophilic three-dimensional networks of crosslinked polymer chains.[20] A desirable trait of hydrogels is their customizability.[20] Different materials lead to unique traits like self-healing through dynamic crosslinks, hydrogen bonds, and ionic bonds as seen in Figure 2.[15,17,23] Hydrogels are commonly used to mimic physical and biochemical conditions found in the human body.[24] They have similar water content and mechanical properties to human tissue.[22] However, conventional hydrogels are fragile, limiting their use in e-skin; as a result, recent research has gone into developing tough hydrogels that are flexible and stretchable.[23] Researchers have used tough hydrogels to create self-healing e-skin.[23]

1.2.1. Materials:

Hou *et al.* created a hydrogel by combining a flexible polymer network and a rigid conductive polyaniline network with ionic and hydrogen bonds.[15] This resulted in a polymer hybrid gel with excellent mechanical toughness, puncture resistance, and self-healing at room temperature.[15] Polyaniline is piezoresistive and has high conductivity, making it an excellent material for e-skin.[15] Due to the self-healing mechanism being based on dynamic hydrogen and ionic bonds, the self-healing is inconsistent in water.[15]

Song *et al.* designed a collagen-based hydrogel using double network theory and coupling noncovalent interactions and dynamic covalent bonds to enhance their e-skin's mechanical and environmental stability.[17] Collagen has the advantage of excellent conductivity, biodegradability, and biocompatibility but suffers from poor mechanical strength; thus, it synergizes exceptionally well with double network theory.[17] In addition, collagen is a primary component of skin, allowing the hydrogel to imitate human tissue.[17]

To further emulate human tissue and bone, Zhang *et al.* chose a wood-based hydrogel for its natural anisotropicity, a unique property found in human muscle, skin, and bone.[26] They hypothesized that the wood's anisotropic structure would allow it to detect more complex signals.[26] They combined a wood fiber skeleton with wood-hydrogels made of polyvinyl alcohol to create an e-skin with anisotropic properties and self-healing capabilities by crosslinking cellulose nanofibers and PVA.[26]

1.2.2. Results and Discussion:

Hou *et al.* tested the strength of the e-skin before and after self-healing.[15] They stretched their sample until it reached a critical strain point – when the sample lost more energy than it could store.[15] Once the sample reached 300% strain, they dropped the strain to 1%, allowing the sample to recover.[15] They found that the e-skin's strength began to recover after reaching a critical strain point, showing that the ionic bonds and hydrogen bonds were reforming without outside stimuli like heat.[15]

Likewise, the collagen-based e-skin also managed to display self-healing properties.[17] The e-skin was broken and managed a self-healing efficiency of 80.3% after 24 hours.[17] This remained consistent through multiple cycles.[17]

The wood-based e-skin did not portray self-healing without external stimuli like its counterparts. After breaking, the sample was soaked in water to allow the PVA molecular chains to crosslink before being immersed in sulfate.[26] It returned to 94% strength, showing higher returns than the collagen-based e-skin; however, researchers found that the e-skin could not heal if the e-skin's wood fiber skeleton snapped.[24] This resulted in a permanent loss of 40% of its mechanical strength.[26] It also portrayed less elasticity than the other e-skins, breaking at 463% elongation.[26] Comparatively, the polyaniline hydrogel e-skin broke at 4000% elongation, and the collagen hydrogel e-skin broke at 900% elongation.[15,17]

Despite the potential upsides of anisotropic materials and wood self-healing hydrogel e-skins, their current downsides of complex synthesis processes and high costs show that they need further development. When focusing primarily on a functional e-skin with self-healing, pursuing an anisotropic material does not appear to be worth the current downsides until further solutions are developed. Collagen is a prime material for further research to develop biocompatible self-healing e-skins. Its main advantage over non-collagen-based e-skins lies in its environmental sustainability.[17]

2. Cost and Sustainability:

One of the biggest challenges of commercializing e-skin for widespread use is the cost and complexity of manufacturing functional e-skin. Traditional polymers for e-skins are often non-renewable, expensive to produce, and cause pollution.[8] Graphene, for example, is commonly chosen for e-skin for its high conductivity, mechanical strength, and flexibility; however, it is expensive to manufacture and can negatively affect the environment.[27,28] In addition, many current e-skins require complex, costly, and time-consuming preparation processes that are not economically viable or environmentally sustainable.8 As a result, researchers have attempted to make the

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manufacturing process simple, cost-effective, and consistent while still producing materials with desirable traits like lightweight, flexibility, durability, and sensitivity.

2.1. Ideas and Proposals:

Researchers turned to paper for cost-effective and sustainable e-skins. A recent study by Chen *et al*. presented an all-paper-based method with carbon-based sensing materials to create a low-cost, ultra-thin, and high-performance pressure-sensitive e-skin.[29] Chen *et al*. chose carbon-based materials – carbon black, carbon oil, and graphite – because of their cost-effective performance as sensors.[29] They hoped that graphite functional layers and carbon black electrodes would be effective in creating a sensitive e-skin at a low cost.[29]

Lai *et al*. also chose to utilize paper and graphite for their e-skin.[30] They utilized paper differently than Chen *et al*. – due to micropores on the surface of paper, paper can function as a nanogenerator with the help of conductive elements like graphite.[30] Thus, they hoped to produce a self-powered, low-cost, and easily manufactured paper-derived e-skin.

However, three drawbacks of paper are its relatively low mechanical strength, inability to stretch, and low durability.[7] To overcome this problem, Zhao *et al*. proposed the creation of a nonwoven fabric-based e-skin using polylactic acid (PLA) and carbon nanotubes (CNT) and a cost-effective ultrasonic dip-coating method.[7] Nonwoven fabric was chosen instead of paper due to its low manufacturing cost and high mechanical strength.[7] In addition, the nonwoven fabric had porous microstructures that were beneficial when integrating CNTs.[7] PLA was chosen for its biodegradability and high hygroscopicity.[30] In addition, PLA is a cheap and eco-friendly plastic.[30] CNTs are also low cost compared to other common alternatives, such as metallic nanowires (MNW) and graphene, while still retaining high carrier mobility, high electrical and thermal conductivity, excellent mechanical properties, and chemical stability despite their tendency to agglomerate.[7,8]

Yang *et al*. also chose to utilize CNTs for similar reasons in their cost-effective e-skin; however, they decided to use chitin as the material for their e-skin because of chitin's high natural abundance, structural stability, remarkable mechanical properties, and favorable biocompatibility.[8] Notably, Yang *et al*. attempted to solve the CNT agglomeration issues by grafting sulfonic acid groups onto their CNTs to make sulfonated carbon nanotubes (SCNT).[8] In theory, the sulfonic acid groups would allow the SCNTs to form a more evenly dispersed and conductive network within the chitin.[8]

2.2. Fabrication Techniques:

The procedures to fabricate the e-skins must be understood to better understand their economic viability.

Despite Chen *et al*. and Lai *et al*. using similar materials, their fabrication processes differed.[29,30] Chen *et al*. used water-soluble graphite to coat paper inside of a mold, before designing and printing carbon interdigital electrodes.[29] The two layers were aligned and sealed with a hotmelt film and heating rod, resulting in a 71 μm thin e-skin.[29] On the other hand, Lai *et al*. used sandpaper as a template to create microstructures with graphite, which was drawn onto the paper in consistent strokes.[30] Then, they assembled the paper by coating the backside with Ecoflex 00-50™ and sealing the papers with tape.[30] Arguably, Chen *et al*.'s water coating method would be more consistent than Lai *et al*.'s method. However, Lai *et al*. reported that their method was highly reproducible and prepared five identical sensors that produced equivalent results.[30]

Zhao *et al*. also produced multiple e-skins with their methodology; notably, each e-skin they produced had a different function.[7] They produced one pressure sensor (P-S), one stretchability, flexion, and temperature sensor (S/F/T-S), and one humidity sensor (H-S).[7] To prepare their e-skin, they used ultrasonic treatments to create an aqueous dispersion of CNTs and immersed the PLA fabric before sonicating it.[7] Then, they baked it in a drying oven.[7] On the other hand, Yang *et al*. sulfonated their CNTs.[8] The process involved various chemicals – such as P-amino-benzenesulfonic acid, sodium hydroxide, sodium nitrate, and hydrochloric acid – and a vacuum heating table.[8] They also used purified their chitin using various bases, acids, and freeze-drying.[8]

2.2. Findings:

The four research groups tested various mechanical aspects of their respective e-skins. Additionally, the raw material costs of major materials in each of the e-skins are reported via Business Analytiq.

Table 1: A summary of data points reported in the previously mentioned studies. Price Indices are sourced from Business Analytiq to provide consistency across data points.

E-Skin Material	Pressure Sensitivity [kPa^{-1}]	Response Time (ms)	Recovery Time (ms)	Ref.	Price Index in NA (Material - USD/kg)
Paper and carbon-based materials	308.54 [0.01-1 kPa]	26 (2 kPa) 8 (10 kPa)	197 (2 kPa) 9 (10 kPa)	Chen *et al*., [29]	Graphite - 1.3, [34]
Paper and graphite materials	7202.2 [0-1 kPa]	25.0	50.1	Lai *et al*., [30]	Graphite - 1.3, [34]
PLA/CNTs (P-S) (S/F/T-S)	19.12 [0.015-6.125 kPa] 5.38 [6.125–12.005 kPa]	151.7 (12.005 kPa)		Zhao *et al*., [7]	PLA - 2.32, [35]
Chitin/SCNTs	1.75 [0-5 kPa]	10		Yang *et al*., [8]	

As seen in Table 1, the paper-based e-skins had much higher pressure sensitivities along their pressure ranges than Zhao *et al*. and Yang *et al*.'s e-skins: 308.54 kPa^{-1} and 7202.2 kPa^{-1} versus 19.12 kPa^{-1} and 1.75 kPa^{-1} respectively.[7,8,29,30] However, the clear outlier is Lai *et al*.'s e-skin, which reported a pressure sensitivity of 7202.2 kPa^{-1} at a pressure range of 0-1 kPa.[30] It also maintained similar response and recovery times compared to its counterparts, being 1 ms faster than the paper and carbon-based materials e-skin at 2 kPa and far faster than the nonwoven PLA/CNT fabric e-skin at 151.7 ms.[7,29,30]

However, the non-paper-based e-skins had advantages over the paper-based e-skins. For example, both Zhao *et al*. and Yang *et al*. reported that their e-skins had elastic properties, while the paper-based e-skins did not report such findings.[7,8,29,30] Zhao *et al*. reported that their e-skin reacted to being stretched, and Yang *et al*.'s e-skin stretched to 10% elongation at its breaking point, giving both far more durability in harsh environments than the paper-based e-skins.[7,8]

It is important to note that the paper e-skins' lack of elasticity limits their viability in scenarios where e-skin must physically adapt to its surroundings, such as on the human body for health-monitoring purposes or during mechanical duress.[31]

Notably, all four studies used carbon-based materials for the sensing and conductive aspects of their e-skins, suggesting carbon may be an optimal material for cost-effective sensing applications. Carbon is the fourth most abundant element in the observable universe and graphite, carbon black, and CNTs are inexpensive materials.[7,8,29,30,32,33] According to Figure 1, graphite is also cheap with a price index of $1.3/kg.[34] Another common trend is using commercially widespread or naturally abundant materials for e-skin – suggesting that natural or commercial abundance is key for cost-effective material approaches to e-skin.

Materials like graphite, paper, and chitin are much more commercially available than CNTs, which typically have complex synthesis methods that require specialized equipment such as arc discharge apparatuses or laser evaporators.[36] Likewise, the fabrication process of Zhao *et al*.'s e-skin required specialized ultrasonic treatment and sonication machinery, which are not readily accessible to most.[7] In contrast, Lai *et al*.'s e-skin required commercially accessible items such as paper, graphite, and Ecoflex 00-50™ and involved a more straightforward fabrication process of graphite being drawn onto the paper to create microstructures.[30] The fabrication process for Chen *et al*.'s e-skin was more complicated than Lai *et al*., featuring a mold, graphite solution, and a printing plate.[29] Nevertheless, both paper-based processes are more accessible than those used for non-paper-based e-skins.

Another factor to consider is the environmental sustainability of the materials used to produce these e-skins. PLA is only biodegradable under industrial composting conditions and anaerobic digestion, which not all cities have proper infrastructure for; under standard conditions, such as in a landfill, researchers have predicted PLA to take one hundred to a thousand years to decompose while releasing methane into the atmosphere.[7,37,38] Paper is another material that contributes to global warming, as it often goes hand in hand with deforestation.[39] Chitin, however, is a natural polymer that is not linked to environmental risks and is highly abundant in nature.[40]

PLA is a mechanically robust material but can potentially harm the environment without proper infrastructure. Paper also threatens the environment and is a weaker material, but paper-based sensors are cheap. The chitin-based e-skin boasts the most environmentally friendly material makeup. If further research produces chitin-based e-skin that performs at a high pressure-sensitivity, chitin should be among the forerunners for producing cheap, affordable, and sustainable e-skin.

Conclusion

Self-healing materials for e-skins and sustainable materials and processes for e-skin development are critical challenges to e-skin development that merit further research. Two kinds of materials, elastomers and hydrogels, are both excellent choices for self-healing mechanisms; elastomers exhibit mechanical strength, elasticity, and flexion. Hydrogels emulate human tissue and offer high biocompatibility, in addition to their potential for self-healing. PDMS elastomers and TPUs are both materials worth investigating further because of PDMS's ability to self-heal in liquid and for TPU's high self-healing efficiency of 91±4% elongation and 108±14% tensile strength. Meanwhile, anisotropic hydrogels warrant further research for their unique structure, which mimes biological tissue like muscle and bone. Collagen-based hydrogels currently show promise for self-healing biocompatible e-skins that can produce consistent self-healing results. That biocompatibility also places hydrogels as premier options for body-compatible e-skin sensors, while elastomers are more applicable for non-human circumstances like robotics.

Paper and carbon-based e-skins offered high pressure-sensitivity performance at a low cost, far outstripping the performance of the non-paper-based e-skins reviewed in this study. Meanwhile, the PLA nonwoven fabric and CNT-based e-skin are mechanically robust. However, neither e-skins are environmentally sustainable due to their contributions to global warming and general landfill waste. For environmentally sustainable e-skin development, researchers should continue to look for naturally abundant polymers like chitin and conduct further research to improve their pressure-sensitivity. Further research will lead to a sustainable path forward for commercial e-skin. Inevitably, the end goal for e-skin is the combination of self-healing, high performance, and mechanical robustness with environmentally and commercially sustainable production to create commercially available high-performance self-healing e-skin.

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