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Research Progress in Stretchable Circuits: Materials, Methods, and Applications

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Stretchable electronic devices, with their excellent properties such as stretchability and conformal contact with complex surfaces, are widely used in health monitoring and human-computer interactions. For stable operation of these devices, stretchable circuits are crucial, enabling integration of sensors, data collection, and transmission while under strain. To achieve stretchable system integration, addressing key issues such as elastic substrates, sensor devices, rigid components, stretchable electrodes, and packaging layers is necessary. The stretchable electrodes, which connect various functional devices, are crucial for achieving stretchability and are the main focus of this review. Firstly, this work explores the enhancement of electrode design to maintain good electrical conductivity even under stretching conditions, through studying stretchable conductive materials and the structural design of conventional materials. This work then presents studies on substrate interfaces and the design of elastic substrates, such as rigid islands, to enhance the stability of stretchable circuits during use. Finally, the article discusses the applications of stretchable circuits, the challenges they face, and provides new research directions and ways to develop stretchable circuits.

1. Introduction

Stretchable electronic devices have many advantages due to their ability to stretch and conform to complex 3D planes, such as the human skin.[1,2] They are superior to traditional rigid devices that cannot be stretched and may cause discomfort when worn. Ongoing research and development are making stretchable electronic devices capable of detecting more bio-signals, enabling new forms of human-computer interaction. These devices are finding applications in various fields, including stretchable displays,[3,4] multifunctional sensing arrays,[5] wearable health monitoring devices,[6] and smart robots.[7]

To achieve stretchable circuits, flexible substrates, functional components, stretchable electrodes, and packaging must be developed. The design of stretchable electrodes and substrates is crucial. The primary issue with stretchable electrodes is their poor conductivity under tensile conditions. Researchers have focused on developing stretchable conductive materials[8] that can maintain electrical conductivity even at large strains, while also exhibiting skin-like mechanical properties[9] such as strength, flexibility, and stretchability.

Various geometries,[10] such as serpentine or origami patterns, can deform these materials when subjected to strain, which helps to solve the problem of electrical failure that occurs with conventional metallic materials.[11] Furthermore, stretchable structures have been designed to be compatible with traditional printed circuit board (PCB) soldering processes and to enable traditional metal materials to have stretching properties.

The electrodes and functional components are separated from the substrate during stretching, due to the significant chemical differences among stretchable electrodes, substrate and functional components. To address this issue, the elastic substrate's interface is chemically treated[12] to reduce the difference between the two interfaces and increase the adhesion of the interface through chemical bonding.[13] This avoids separation of the electrode from the flexible substrate during operation, which can lead to electrical failure. Large differences in mechanical properties between non-stretchable commercial electronic components and stretchable electrodes cause stable connections between them to be problematic. A strain isolation strategy can effectively solve the difference in mechanical properties between electrodes and components, allowing them to maintain a stable connection under strain and provide protection for non-stretchable components.[14]

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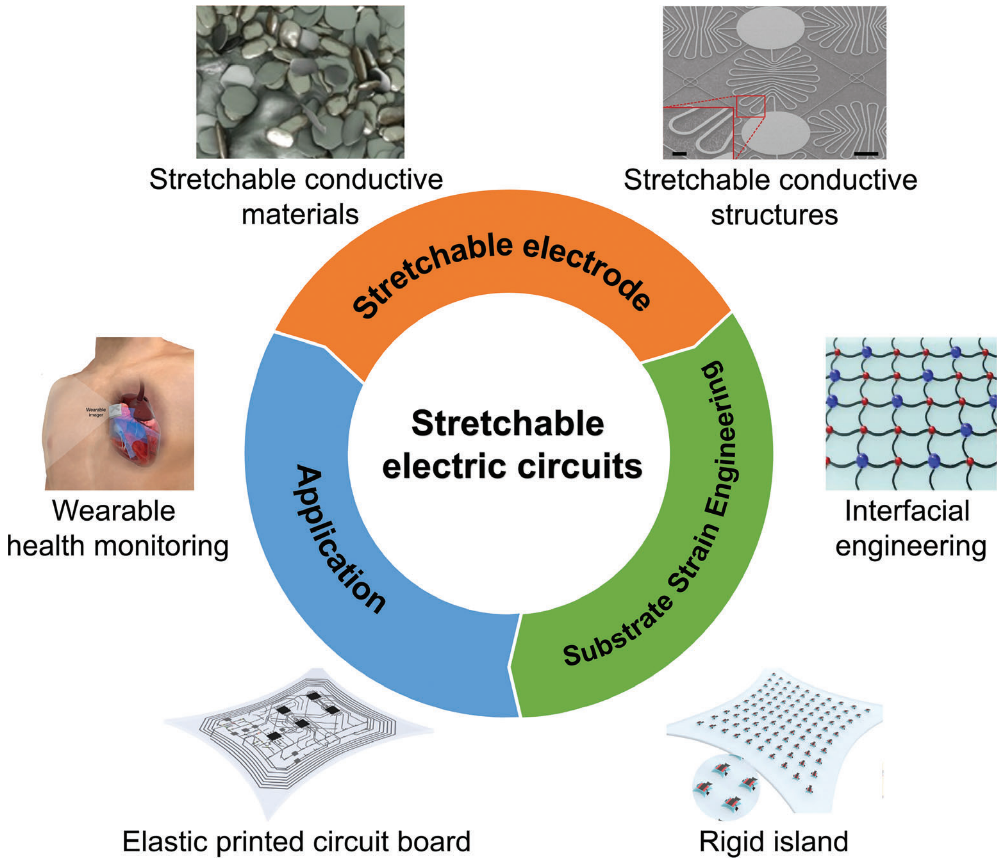

Figure 1. Design and applications of stretchable circuits. Reproduced with permission.[18] Copyright 2021, American Chemical Society. Reproduced with permission.[19] Copyright 2018, Nature Publishing Group. Reproduced with permission.[20] Copyright 2022, Nature Publishing Group. Reproduced with permission.[21] Copyright 2021, Nature Publishing Group. Reproduced with permission.[22] Copyright 2022, American Association for the Advancement of Science. Reproduced with permission.[23] Copyright 2023, Nature Publishing Group.

The development of stretchable electrodes has provided stability in the development of stretchable circuits, promoting potential applications in wearable health monitoring,[15] prosthetics,[16] and soft robotics.[17] In this article, we discuss the development of stretchable circuits in five chapters, as shown in **Figure 1**. Chapter 2 introduces various types of stretchable conductive materials, including liquid metals with self-healing effects, fabrics and fibers with high comfort levels, and stretchable conductive inks compatible with proven printing technologies. Chapter 3 focuses on the structural design of traditional metal materials, such as origami structures, kirigami structure, and serpentine structures, to achieve stretchability. Chapter 4 discusses the substrate for interface treatment and the design of rigid islands to increase the stability of stretchable circuits during use. Chapter 5 discusses the applications of stretchable circuits in areas such as flexible printed circuit boards and health detection. We also address the challenges faced in the development of stretchable circuits and provide new directions and avenues for their application. Overall, the development of stretchable circuits offers great potential for the advancement of electronic devices that can conform to the body's various shapes, sizes, and movements, with potential applications in wearable health monitoring, prosthetics, and soft robotics.

2. Stretchable Conductor Materials

The development of stretchable conductive materials is an exciting area of research for stretchable electrodes as they provide both stretchability and conductivity, enabling the preparation of fully stretchable electronics when connected to other stretchable functional devices such as stretchable transistors and stretchable displays. In this section, we will discuss different types of stretchable conductive materials, including fabric materials that are comfortable and breathable, inks that can print metallic materials that are well compatible with existing devices such as inkjet printing, and liquid metals that exhibit good stretchability and self-healing effects.

2.1. Liquid Metals

Liquid metals have become a popular choice for researchers in stretchable circuits due to their low melting point, good mechanical stretchability, thermal conductivity, and electrical conductivity.[24,25] Moreover, their ability to flow or change shape into various forms at room temperature has made them a promising material to resolve the electrical failure problem that occurs in conventional materials with conductive materials under stretching conditions.[26,27] While research on liquid metal originates from Hg, which have been eliminated due to its high toxicity and volatility, EGaIn (75.5 wt% Ga, and 24.5 wt% In) and Galinstan have been widely used in stretchable point circuit. Their conductivity is 3.4×10^6 and 3.5×10^6 S m^{-1}, respectively, which is 1/20th of copper's conductivity but greater than common stretchable conductive materials. Additionally, when liquid metal is exposed to air, it forms an oxide layer to promote the combination of the metal and other materials. The oxide layer of the liquid metal flows down to the surface of the newly created elastomer under tensile conditions.[28] When the elastic substrate is stretched, the droplet of liquid metal deforms accordingly, increasing the contact area with the elastic substrate. The liquid metal layer deforms with the stretched elastic substrate, and after releasing the strain, the oxide layer rebuilds its surface according to the shrinking elastomer, reducing the contact area between the liquid metal and the elastomer by increasing the thickness of the liquid metal layer and shortening the length (**Figure 2**A). As a result, liquid metals are less susceptible to large resistance changes at higher stretching.

Recent methods of preparing liquid metal for stretchable circuits include direct-write extrusion,[29] micro-contact printing,[30] inkjet printing,[31] selective wetting,[32,33] and masked deposition.[34,35]

Among these methods, inkjet printing technology is extensively used in the research of stretchable circuits due to its high degree of automation, fast preparation speed, and the advantages of high resolution. Liquid metals are fluid in the atmosphere resulting in good compatibility with inkjet printing, it is possible to precisely deposit liquid metals on the substrate. Mahmoud Tavakoli and his cooperators developed a duplex Ag-In-Ga ink with high conductivity and stretchability that can be extruded and printed with high resolution in a manner similar to inkjet printing of granular conductive materials (Figure 2B). The ink consisted of EGaIn, Ag microsheets, and styrene-isoprene block copolymer was synthesized to obtain ultrathin multilayer circuits with high stretchable (the maximum strain of 600%) and excellent conductivity (7.2×10^5 S m^{-1}).[18] The resistance of the circuit only grows from 1.6 to 3.1 Ω under 100% strain, and there is no significant change in resistance over 1000 cycles.

The wettability difference between the liquid metal and the elastomer, shown in Figure 2C, is due to the intermetallic compounds formed at their interface. Desheng Kong and coworkers patterned liquid metal conductive pathways by coating liquid metal on patterned Cu films in a dilute hydrochloric acid solution.[32] The naturally occurring oxide layer protects the liquid metal's structural integrity, and the resulting patterned circuit showed a resistance increase of only threefold at 100% strain. It maintained this resistance within 1000 tensile release cycles at 300% strain, indicating its excellent electromechanical durability.

Mahmoud Tavakoli and his team proposed a strategy for preparing a GaInSn-based circuit by smearing eutectic GaInSn alloy on a pretreated substrate spin-coating with polyvinyl alcohol (PVA) and inkjet printed with silver nanoparticle ink.[36] The circuit's conductivity was 6.65×10^6 S m^{-1}, and it maintained good conductivity under 100% strain (Figure 2D).

Direct patterning of pure liquid metals into high-resolution circuits is challenging due to their surface tension and mobility, which cause them to gather into low surface energy spheres during processing.[37] Additionally, the poor adhesion of liquid metals to many common substrates, such as textiles, limits their applications. Therefore, researchers typically modify liquid metals during the liquid metal research stage. Figure 2E depicts a recyclable liquid metal micro-gel ink proposed by Yong He's group, which encapsulates liquid metal in an alginate micro-gel shell. During mechanical stirring, Ga ions from the liquid metal crosslink with sodium alginate to form a micro-gel that adheres to the liquid metal surface. The ink exhibits shear-thinning properties via hydrogen bond formation and breakage under different stress conditions,[38] providing excellent printing adaptability and adhesion to various substrates. Moreover, this ink enables direct printing of functional circuits on clothing to prepare and study electronic clothing.

Liquid metals are usually non-conductive in their initial state, and their conductivity requires disrupting the oxide shell around liquid metal nanoparticles via micro-strain, pressing, freezing and ultraviolet laser beams. Regardless of the patterning method, an encapsulation or additional injection step is necessary for all practical end applications to mechanical wear or surfaces contact from damaging the liquid metal circuit.

2.2. Fibers/Textiles

Stretchable circuits are typically integrated onto a flexible substrate, but to ensure the stability of the circuit operation while providing wearing comfort,[39] elastic fibers/textiles with a porous network,[40] high specific surface area and low Young's modulus[41] are needed. To protect the skin and maintain a breathable environment, it's crucial to develop stretchable circuits that allow the exhaust of body fluids. Textiles, which are porous structures composed of flexible materials, must withstand mechanical, thermal, and chemical corrosion during daily activities and washing.[42] Direct coating of textiles with conductive materials is a simple and scalable route[43] that can be achieved with existing production processes. Wojciech Matusik and his co-workers developed an automated coating technology to wrap up fibers with piezoresistive nanocomposites to obtain coaxial piezoresistive fibers[44] that were integrated with industrial digital machine weaving technology to turn functional fibers into large-scale textiles with arbitrary 3D geometry. Kinam Kim and his team immersed electrospun poly(styrene-*block*-butadiene-*block*-styrene) (SBS) fibers into a precursor solution of silver nanoparticles, where in situ conversion of silver nanoparticles occurs on the fibers, maintaining a high bulk conductivity of 2200 S cm^{-1} at 100% strain[45] (**Figure 3**A).

However, printing/coating functional materials such as conductive fillers onto textiles is prone to problems such as cracking and delamination due to the mechanical mismatch. Conductive materials coated on textiles can stiffen the fabric and clog the porous structure, required comfort. To solve these issues, methods such as direct fibrillation preparation of conductive composites or embedding of conductive functional materials into textiles have been proposed. Electrostatic spinning is a simple, versatile, and economical method that can obtain large high specific surface area and porosity, improving the comfort of people for daily use.[46] Yury Gogotsi and his team used

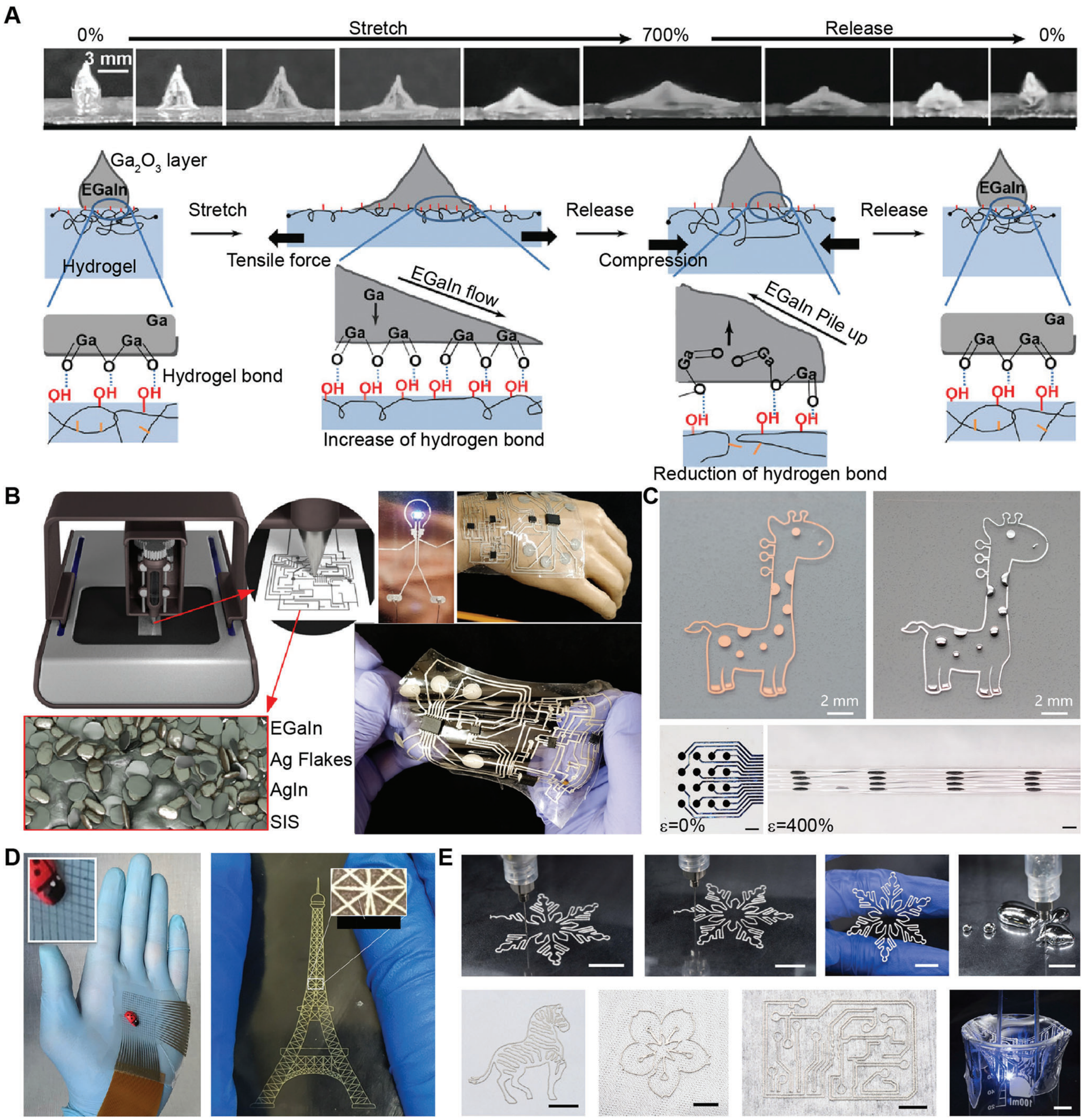

Figure 2. The method to achieve patterned liquid metal. A) Exploration of the working mechanism of liquid metal to maintain conductivity under stretching conditions. Reproduced with permission.[28] Copyright 2020, Wiley-VCH. B) Printing liquid metallic ink via extrusion printer. Reproduced with permission.[18] Copyright 2021, American Chemical Society. C) Patterning via selective wetting with liquid metal. Reproduced with permission.[32] Copyright 2022, American Association for the Advancement of Science. D) Inkjet printed stretchable circuits. Reproduced with permission.[36] Copyright 2020, Wiley-VCH. E) Inkjet printed stretchable circuits. Patterned printing of liquid metal inks on various substrates can be realized by modifying liquid metal. Reproduced with permission.[38] Copyright 2022, American Chemical Society.

electrostatic spinning to obtain nanofiber films with MXene content of 35 wt% (Figure 3B), improving the electrical conductivity of the nanofibers, and embedding the Mxene into the nanofibers without binders or additives.[47] The preparation of composite electrodes solves the problem of stripping the conductive material from the substrate during folding and bending, improving the electrode's durability. Although electrostatic spinning technology enables the preparation of fibers with electrical conductivity, the current composite solution containing conductive particles can lead to agglomeration or stacking of particles during

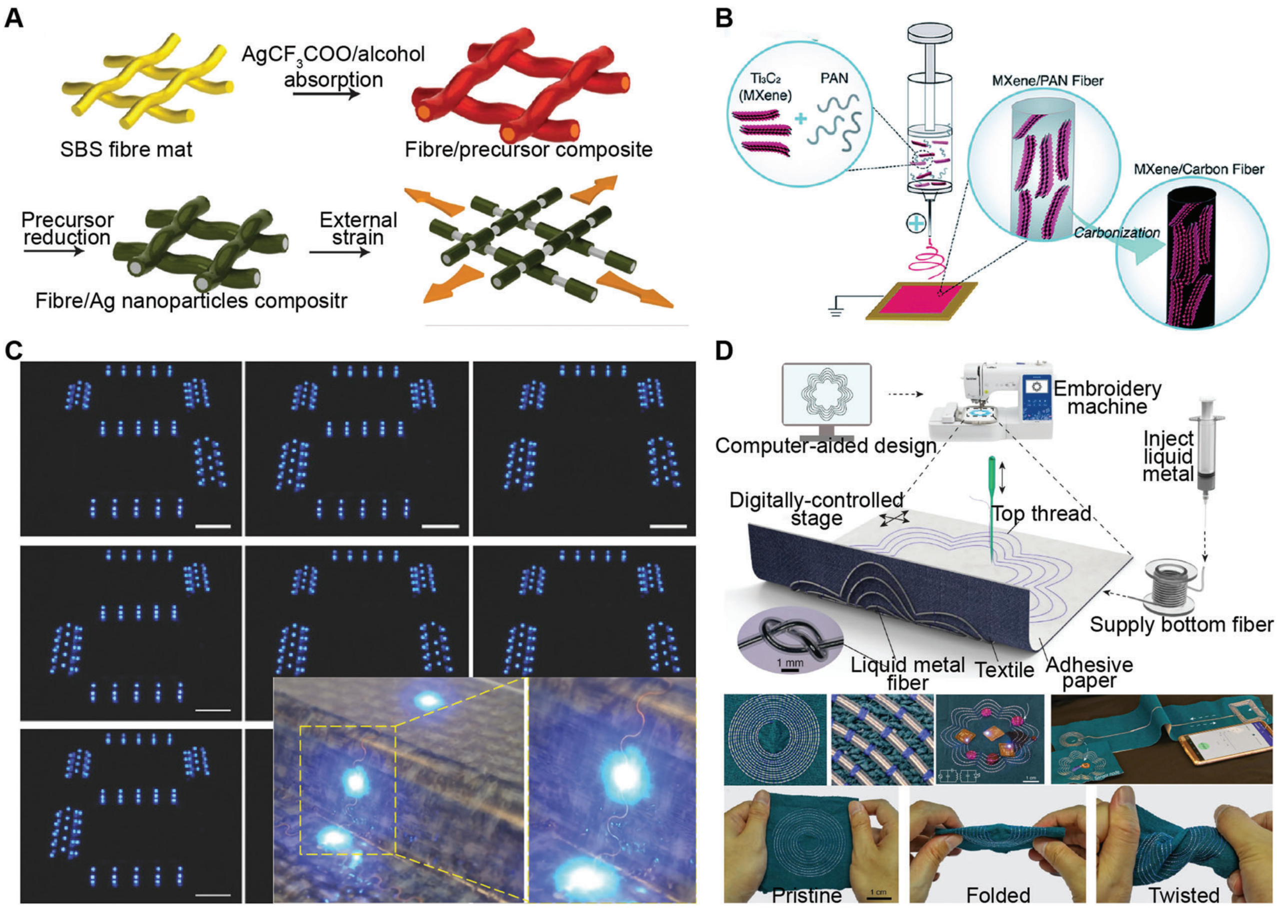

Figure 3. Preparation of stretchable conductive fibers/textile. A) Direct coating of conductive materials on electrospun fibrous membranes. Reproduced with permission.[45] Copyright 2012, Nature Publishing Group. B) Preparation of conductive fiber membrane by direct electrospinning of mixed conductive filler and polymer. Reproduced with permission.[47] Copyright 2019, Royal Society of Chemistry. C) Transfer of conductive pathways to fabric by lamination. Reproduced with permission.[49] Copyright 2018, Wiley-VCH. D) Conductive fibers are embedded into the fabric through the embroidery process. Reproduced with permission.[50] Copyright 2022, Nature Publishing Group.

electrostatic spinning, forming beads on the fibers, thus affecting the homogeneity and electrical conductivity of the fibers.[48] Jan Vanfleren and his team developed a snake-shaped stretchable structure electrode by transferred a stretchable circuit onto a textiles (nylon) using a lamination process.[49] The process involved bonding melted nylon (PA6) to pre-prepared stretchable circuits and electronic components, resulting in a stretchable electronic display that could display different symbols through a control circuit (Figure 3C). However, this process required operation at the glass transition temperature of the fabric material, which damaged the original porous structure of the fabric. In contrast, the embroidery process maintained the same porous structure of the conductive pattern and substrate textile, allowing the conductive pattern to breathe as well as the substrate textile, and has good compatibility with commercial embroidery equipment.[44] John S. Ho and colleagues demonstrated a method for preparing liquid metal fibers by infiltrating liquid metal into Perfluoroalkoxy (PFA) tubes and bonding them to clothing by embroidery (as shown in Figure 3D). Due to the high electrical and mechanical properties of liquid metal fibers and their compatibility with digital embroidery processes, complex patterns could be designed by established computer-prepared textile processes.[50] Conductive thread embroidery maintained the flexibility and permeability of textiles and could be produced in a scalable manner using existing manufacturing technologies. The resulting conductive textiles demonstrated good stability, with less than 3% change in relative resistance after simulated daily wear conditions, including 24 000 cycles in a phosphate buffer saline (PBS) solution at 50 °C and more than 10 h of machine washing.

2.3. Metal Nanoparticle Printable Conductive Inks

Due to their advantages of simple preparation, high pattern resolution, and low production cost, processes such as screen printing,[51] inkjet printing,[52,53] stencil printing,[54] and direct nozzle writing[55,56] are commonly used for stretchable circuits. However, liquid metal as a printable ink poses challenges due to its high surface tension and potential for leakage, while commercial printing inks suffer from poor stretchability, low adhesion to elastic substrates, and high resistivity (higher than bulk metal).[57] To address these issue, researchers have focused on developing new printing inks.

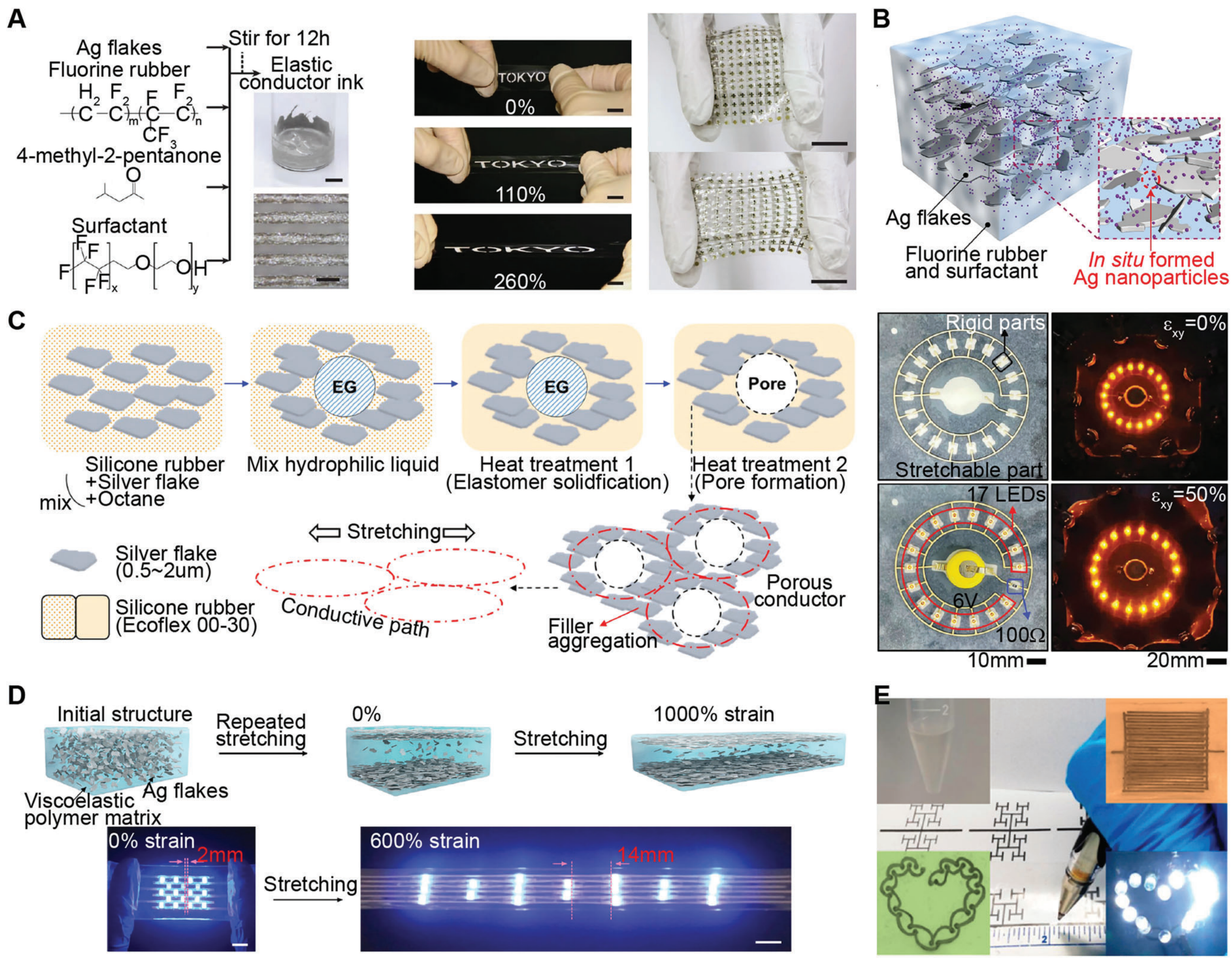

Figure 4. Preparation of stretchable conductive ink. A) Adding surfactants to enhance the adhesion between silver ink and substrate. Reproduced with permission.[58] Copyright 2015, Nature Publishing Group. B) In situ generation of secondary microstructures in silver inks. Reproduced with permission.[60] Copyright 2017, Nature Publishing Group. C) Formation of hollow microstructures in silver ink keeps conductive pathways intact during stretching. Reproduced with permission.[61] Copyright 2023, Wiley-VCH. D) Induced self-assembly of silver ink. Reproduced with permission.[62] Copyright 2022, Wiley-VCH. E) Handwritable conductive ink. Reproduced with permission.[63] Copyright 2016, American Chemical Society.

Takao Someya's team developed printable elastic conductive inks using silver flakes, fluoroelastomers, and fluorosurfactants, as shown in **Figure** 4A. The surfactant modifies the surface of silver flakes, increasing their affinity with fluororubber and enabling a conductivity of 182 S cm^{-1} under 215% stretching. Organic transistor integrated circuits were fabricated on elastomers with modulus gradients by stencil printing and could work stably at large strains of 130%.[58] Building on previous work,[58,59] Naoji Matsuhisa developed a conductive ink that forms silver nanoparticles in situ, as depicted in Figure 4B. This ink, which mixes micron-sized silver flakes, fluoroelastomer, and surfactants, can achieve a conductivity of 6000 S cm^{-1} in the initial state and maintain a conductivity of over 900 S cm^{-1} under a strain of 400%.[60] By fully printing an elastic sensor network using this conductive ink, Matsuhisa demonstrated high compatibility with high-throughput printing processes.

Sungmook Jung et al. developed a novel method for creating a hollow-like internal structure in silver conducting particles using a hydrophilic solution (Figure 4C). By connecting silver nanoflakes in a circular arrangement along the porous structure, they were able to form conducting pathways that remained constant even after structural deformation during stretching.[61] Importantly, the electrical resistance of the conductive material did not change with increasing strain. To demonstrate the practicality of their silver ink in stretchable electronics, they constructed a circular circuit by connecting a cell, light-emitting diode (LED), and resistor with nozzle-printed electrodes. The circuit was able to operate properly at 50% repeated strain, with the brightness of the LED remaining unchanged. Hangxun Xu and his co-workers addressed the issue of increasing distances between conductive fillers under tensile conditions, which can lead to blocked charge transport,[62] as shown in Figure 4D. They achieved a higher surface conductive path by mixing silver flakes with elastic substrates composed of polyethylene glycol and polyaniline. These components can form hydrogen bonds with each other, which dynamically break and recover under repeated stretching to consume mechanical energy. The prepared conductive ink was then printed onto an elastomer by screen printing to obtain a complex stretchable circuit. A stretchable LED array was created by connecting the LED, and the array worked stably at 600% strain while maintaining a stable light intensity.

Writing has developed into an important method for manufacturing various circuits and functional patterns with its simple advantages. Jun Yang's group developed a composite conductive ink that can be directly loaded into a ballpoint pen, allowing for the creation of different patterns through direct writing and manual copying (Figure 4E). The composite ink is mainly composed of soluble silver salt, adhesive rubber, and low-toxicity butanone.[63] The ink can be prepared into a particle-free transparent solution due to the high solubility of silver salt in butanone and the salt-assisted dissolution of rubber. The presence of viscous rubber allowed the written conductive ink to maintain good adhesion on various substrates and maintain good conductivity under various deformations. The presence of silver salt in the ink allowed it to be converted into highly conductive silver nanomaterial after in situ reduction. Printable conductive inks are created by dispersing conductive nanoparticles in polymers in various ways. As the polymer substrate is an insulator, the conductivity of the ink depends on the conductive pathways formed by the nanoparticles.[64] While silver nanoparticles in the ink are expected to make direct contact, reducing interface contact resistance, a portion of the silver nanoparticles remain separated within the ink, potentially leading to performance issues.[65]

However, a portion of the silver nanoparticles remain separated within the ink, leading to potential performance issues.[66] Industrial production of printable conductive inks using a specific process remains a major challenge for cost-effective manufacturing. Overcoming these challenges will be essential in realizing the full potential of printable conductive inks for the development of stretchable circuits and other advanced applications.

3. Stretchable Conductive Structure

Compared to stretchable conductive materials, metals such as gold, silver, and copper have high conductivity and are compatible with standard PCB bonding processes. However, these metallic materials are less stretchable and tend to fracture at lower strains, which can lead to electrical failure. To address this issue, a structural design approach has been developed that uses non-stretching rigid materials to maintain high electrical conductivity under strains. This method employs geometric engineering techniques, including serpentine structures, origami/Kirigami structures, 3D spring-like spiral structures, and more. These designs have been proven effective in solving the problem of non-stretchable metallic materials while maintaining high electrical conductivity.

3.1. Origami Structure

Origami is a technique that allows for global stretchability and deformation of rigid or non-stretchable materials by performing folding and bending operations. This enables origami structures to be designed in various sizes and shapes, with stretchability that can be adjusted to specific needs. When applied to stretchable circuits, 2D planes can be transformed into 3D stretchable structures that are compatible with different materials.

Jong-Hyun Ahn was inspired by the 3D deformable structure of the origami process and successfully created stretchable displays that maintained the same pixel density and image quality as the original image when stretched. The stretchable display is composed of a 7×7 light-emitting diode (LED) pixel array integrated into a transparent epoxy resin frame (**Figure 5**A). The Micro-LED chip made of GaN epitaxial wafers, is integrated onto a substrate with a 3D shape and connected with graphene and Au electrodes with good conductivity. The pixels of the stretchable display are hidden in the unstretched state and exposed in the biaxially stretched state, thereby providing high pixel density and quality under 100% stretch.[67] Hongyu Yu transferred flexible conductive polyester electrodes onto a paper base, then folded them according to a pre-designed pattern to form a Barreto's Mars fold structure.[68] A stretchable paper-based folded humidity sensor based on this structure can convert Barreto's Mars folds from a 3D structure to a 2D plane during strain application. This ensures that the humidity sensor is insensitive to applied tensile and bending strains. Origami structures increase the application of non-stretchable paper-based materials, and the humidity sensor can be sewn onto masks, similar to fabrics for detecting people's breathing. While origami structures can achieve intricate and fine designs, they require precise operations and involve many folding curves. Any mistake in the folding process could lead to the entire structure failing. When origami was considered as stretchable structures applied in non-stretchable materials, the strength and stability of the substrate must be carefully considered. Attention to detail is critical during the design phase

3.2. Kirigami Structure

Kirigami structures are similar to Origami but with a distinct difference: they are shear-based folding techniques that offer higher stretchability than Origami structures. Unlike Origami, Kirigami structures can be folded and unfolded in a variety of ways, allowing for a wider range of deformations.[69] Additionally, the preparation method for Kirigami structures is simpler and more versatile than that of Origami, with the possibility of computer-aided design automation.

In the field of stretchable electronics, Lizhi Xu's group has leveraged the potential of Kirigami structures to prepare a stretchable multifunctional sensing array with a kirigami structure that can conform to the skin (Figure 5B). The structure of the paper cut was obtained by laser cutting of the fiber film, resulting in a high stretchability and good compliance on the 3D surface.[70] The sensing array features high-performance sensors and breathable nanofiber substrates that were integrated through a stamp-based transfer technique and electrospinning technology. The circular edge design of the paper-cutting incision reduces stress concentration in the stretching process, resulting in good air permeability and conformal contact with the complex surface of the human body, enabling the monitoring of various physiological parameters.

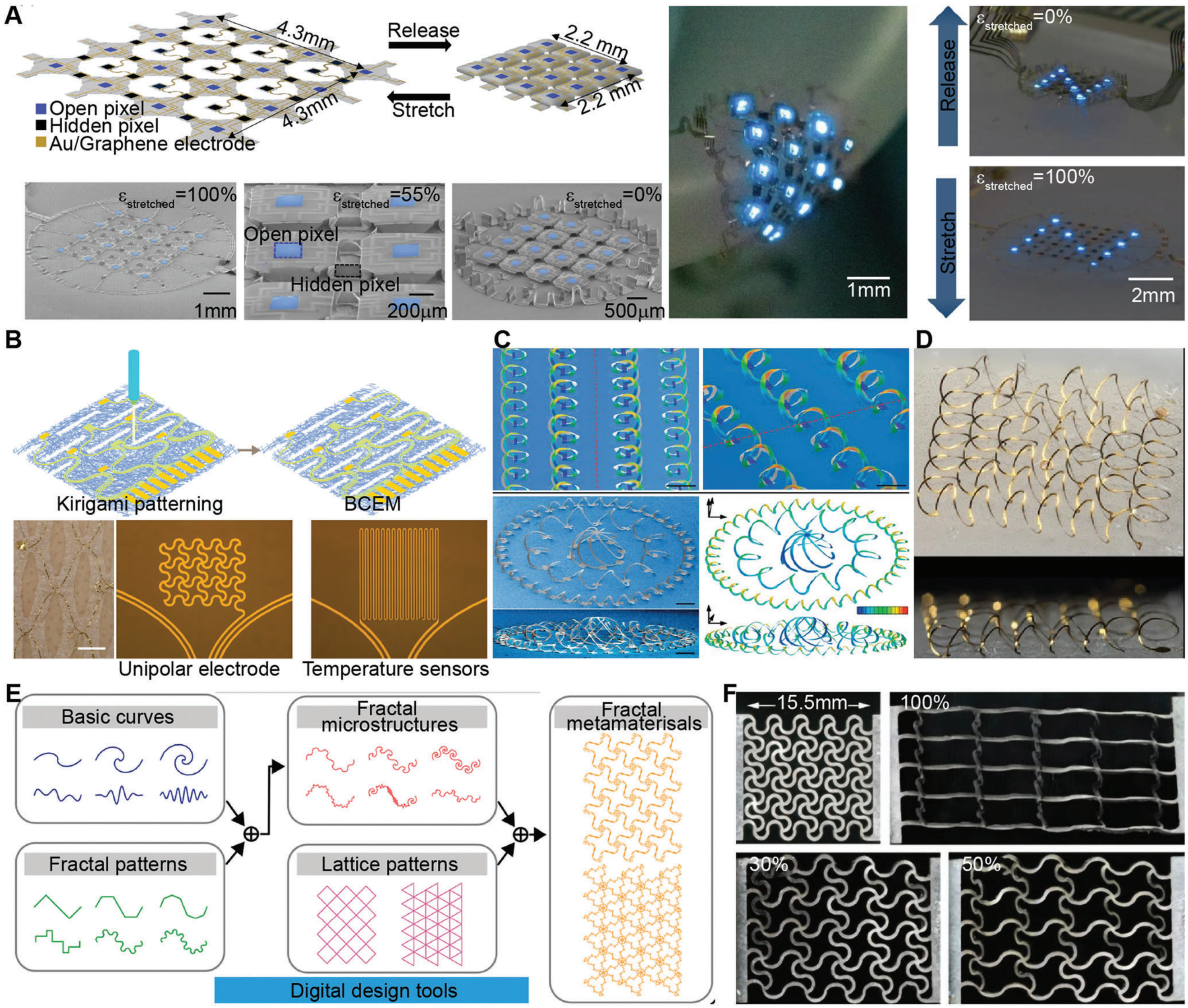

Figure 5. Stretchable conductive structure. A) Origami structure. Reproduced with permission.[67] Copyright 2022, Elsevier. B) Kirigami structure. Reproduced with permission.[70] Copyright 2022, Wiley-VCH. C,D) 3D spring-like spiral structure. Reproduced with permission.[72] Copyright 2015, American Association for the Advancement of Science. Reproduced with permission.[73] Copyright 2017, Nature Publishing Group. E,F) Serpentine structure and fractal structure. Reproduced with permission.[74] Copyright 2023, Wiley-VCH. Reproduced with permission.[75] Copyright 2023, Wiley-VCH.

In addition, Han-Ki Kim's research team combined the stretchable bionic structure of snake skin with kirigami techniques to obtain the best structural design and arrangement through finite element analysis. The layout of the snakeskin structure enables compression along the strain direction ensuring that the resistance of the structure does not change during stretching.[71] This method is ideal for the fabrication of temperature-sensitive materials on electrodes that can detect changes in human body temperature without being affected by strains generated by human motion.

While Kirigami structures exhibited many advantages over Origami, there are still limitations that need to be considered. For instance, notches in Kirigami structures can lead to stress concentration and reduced stability. Therefore, careful monitoring of the applied strain is required to prevent failure of the structure during the stretching process. Despite these limitations, Kirigami structures remain a promising technique with significant potential for use in various applications.

3.3. 3D Structure

The 3D spring-like spiral structure comprises a tightly coiled ring around a central axis that forms a helical structure extending outward from the center. The precise and intricate arrangement of conductive threads enables the 3D spring-like spiral to withstand greater tension and compression without compromising its structural integrity.

Microfabrication technology was first proposed by John A. Rogers' group to create the 3D construction of non-stretchable conductive materials. This involved using a pre-stretched substrate to prepare a snake-shaped stretchable structure, with the lithography process controlling the specific position.[72] Releasing the strain from the pre-stretch resulted in good contact with the substrate, causing bending and buckling at other positions, ultimately forming a 3D spring-like spiral structure (Figure 5C). Further improvements included designing small discs at both ends of the electrode, forming strong covalent siloxane bonds with the substrate of siloxane under biaxial pre-stretching conditions (Figure 5D). Finally, a 3D spring spiral structure was obtained through controlled buckling deformation. Finally, a 3D spring spiral structure was obtained through controlled buckling deformation.[73] The proposed method is compatible with the adaptability of micro-nano technology and the stretchable preparation of rigid non-stretchable materials.

Macroscale spring structures have also been extensively studied.[76] Yong Zhao's team sprayed carbon nanotubes on polyurethane (PU) fibers, and then twisted them to make the fibers form a helical structure.[77] This helical fiber can solve the interfacial bonding problem between carbon nanotubes and PU to form a more stable conductive network. The prepared stretchable fibers were able to maintain their electrical conductivity under high strains of 1700%.

The preparation and processing of 3D spring-like spiral structures require specialized technology and equipment, leading to higher production costs. Careful selection of materials and processes is necessary for optimal performance in practical applications. Despite these challenges, the potential benefits of 3D spring-like spiral structures make them a promising area for research and development in various applications.

3.4. Serpentine Structure and Fractal Structure

The fabrication of stretchable structures, such as origami, kirigami, and 3D helical structures, presents challenges due to their requirement for pre-stretching or laser cutting pretreatment processes and their 3D spatial deformation when subjected to strain. Bending structures, such as the serpentine structure, are commonly used in stretchable conductors due to their ability to deform in-plane under re-tensile strain and their integration with the industrial PCB process.[78,79]

Hee-Tae Jung used conventional secondary sputtering technique to fabricate a high aspect ratio serpentine metal structure that has strain-insensitive characteristics and low strain gradient due to encapsulation in PDMS. The sheet resistance of 7.6 Ω^{-1} can still be maintained under 200% strain, and the transparency does not change during the whole stretching process.[80] Due to the high aspect ratio of its structure, as well as encapsulation in PDMS, the structure has low strain gradient in addition to its strain-insensitive characteristics, which can reduce the phenomenon of strain concentration and strain failure, thereby improving the stability of the structure sex and reliability.

However, the serpentine structure has low integration density, which has led to the development of the layered fractal structure.[11] Guoying Gu and colleagues used theoretical calculations and 3D printing technology to prepare various fractal structures,[74] which can achieve greater deformation than the serpentine structure without mechanical failure due to their deformation in a multi-order expansion manner under strain tension (Figure 5E). By designing the fractal structure and integrating it with LED, it can operate normally within the designed strain. Desheng Kong and colleagues designed a flexible circuit with a power supply using laser ablation of conductive materials and integrated it with an electronic patch, which can control the treatment of wearable cancer through mobile phones (Figure 5F).[75]

Despite the maturity of the preparation processes and theoretical support for stretchable structures, their limited stretchability, inefficient area coverage, and limited mechanical integration still require improvement. Future research should focus on improving these limitations to make stretchable structures more practical and effective in various applications.

4. Substrate Strain Engineering

Current research on stretchable conductive materials and structures has made significant progress in meeting the demand for stretchable electrodes that maintain high conductivity under stretching conditions. However, to ensure the stability of stretchable electrodes during operation, a stable connection must be established between the electrodes, stretchable substrates, and functional devices. Despite recent advancements, there are still challenges to be addressed in stretchable electrode materials. On one hand, the mechanical and chemical differences between the stretchable electrode and substrate can lead to interfacial separation during stress, rendering the electrode invalid. On the other hand, the stable connection of the soft-hard interface between stretchable electrodes and rigid components remains a challenging problem when designing stretchable circuits/systems with rigid silicon-based components. To overcome these challenges, it is proposed to increase the interfacial adhesion between electrode materials and stretchable substrates to reduce interface delamination. Additionally, the design of a rigid island can achieve a stable connection of the soft-hard interface between the stretchable electrode and rigid element under strain.

4.1. Engineering the Interfacial Properties

Unyong Jeong and his colleagues have developed a method to accurately control crack density and depth on elastic substrates by depositing a patterned aluminum layer on a flexible substrate.[81] This technique enables the creation of a tensile conductive transparent gold conductive network through the generated cracks, as shown in **Figure 6**A. The gold conductive network is only present in the crack, and it folds in small strains while expanding under high strains, allowing the network to be displayed on the surface. As a result, the brightness of the LED remains stable under various 100% strains, which is significantly greater than the failure strain of traditional crack sensors (≈2%).

Due to the mechanical mismatch between the metal film and the elastic substrate, the metal film is prone to cracks under strain conditions,[82] and the gold film will fail electrically under smaller strain.[83] To improve the toughness of the interlayer interface and inhibit the generation of cracks under delamination and strain, Zhenan Bao's group has

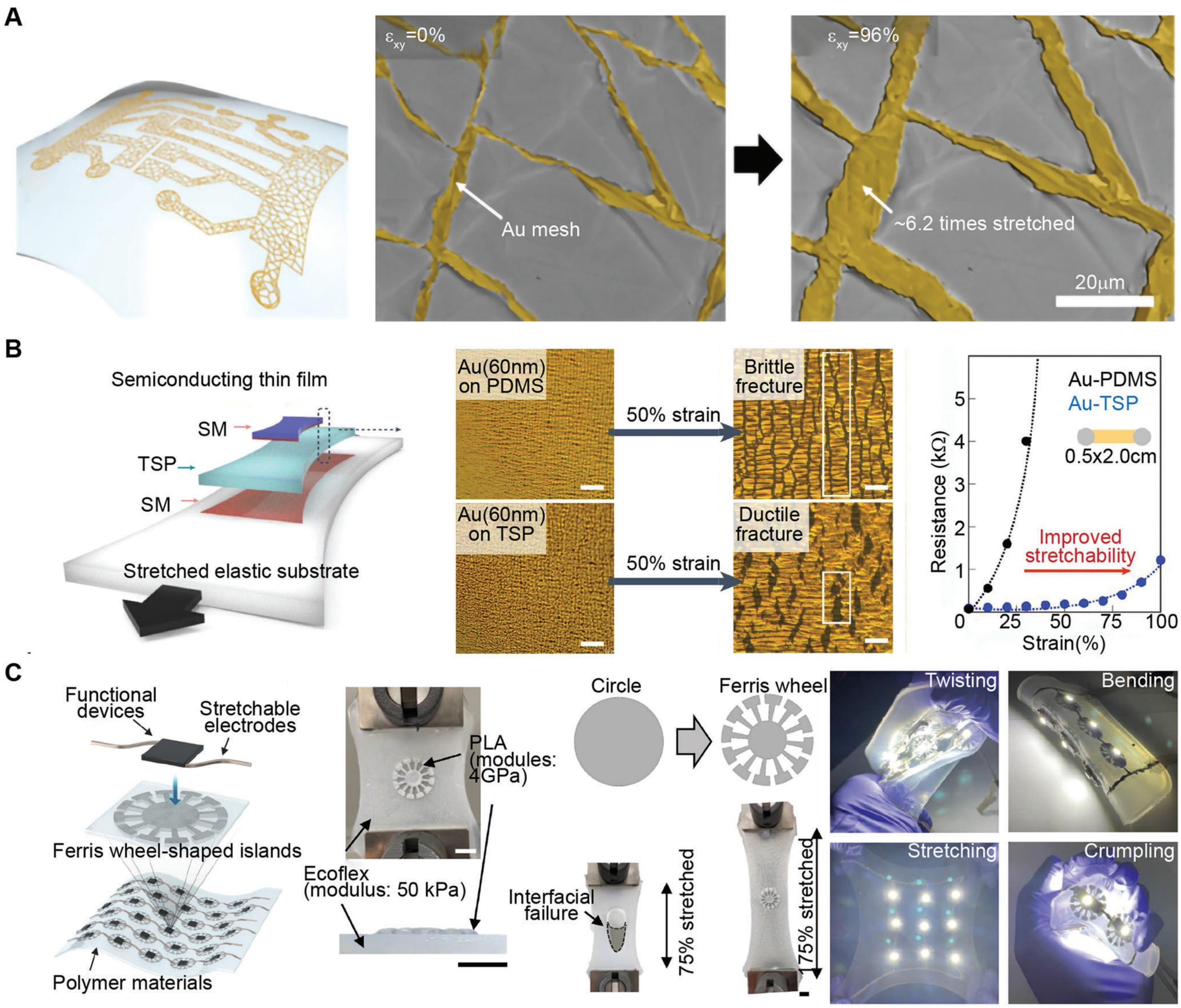

Figure 6. Design stretchable substrates. A) Cracks produced by pre-stretchingand embedment of metal into cracks. Reproduced with permission.[81] Copyright 2021, Wiley-VCH. B) An adhesive layer added between the flexible substrate and the conductive layer. Reproduced with permission.[20] Copyright 2022, Nature Publishing Group. C) Embedment of rigid islands on flexible substrates to protect non-stretchable components. Reproduced with permission.[89] Copyright 2022, American Association for the Advancement of Science.

designed a tough interface between the conductive film and elastic substrate,[20] as shown in Figure 6B. This design incorporates a tough self-healing polymer and a surface modifier between the two interfaces. The self-healing polymer dissipates energy through chemical recombination and recombination during the stretch process, while the surface modifier forms both covalent and non-covalent bonds with the tough self-healing polymer or covalent bonds with elastic substrates. This tough interface changes the gold film's brittle fracture to a ductile fracture under PDMS stretching, greatly improving the gold film's tensile properties.

Despite these advancements, there is still much work to be done in engineering interfacial properties to ensure the stable connection between stretchable electrodes, stretchable substrates, and functional devices.

4.2. Rigid Island

Integrating rigid electronic components with flexible substrates is a simple and practical method due to the multifunctionality and robustness of commercial rigid components.[78] However, the large difference in mechanical properties, such as Young's modulus, between the rigid component and the elastic substrate can cause electrical failure due to the separation of soft and hard materials under tensile conditions.[84] To solve this problem, researchers have developed a substrate strain isolation strategy by placing non-stretchable electronic components on a rigid island.[85,86] Under strain, the surrounding elastomeric part with lower Young's modulus stretches, reducing the strain applied to the rigid component and protecting it.[87] This method enables the integration of commercially available rigid electronic components, such as thin-film transistors, integrated circuits, and printed circuits with stretchable devices, resulting in multifunctional, high-performance stretchable circuits.[88]

Steve Park and his team designed a Ferris wheel-shaped rigid island structure that effectively improves failure strain and service life compared to traditional circular and square rigid islands (Figure 6C). The repeated interlocking structure of the Ferris wheel rigid island with the surrounding flexible substrate of low Young's modulus effectively solves the interface mismatch between the rigid island and the surrounding elastic substrate during the stretching process.[89] The LED and battery pack are placed on the rigid island and printed with an intrinsically stretchable silver electrode, resulting in a stable and normally working array of battery-assembled LEDs under deformation conditions such as stretching, twisting, and bending.

Zhenan Bao's team prepared regions with different Young's modulus on the same elastic substrate by changing the crosslink density of the elastomer.[90] Placing non-stretchable transistors onto the elastomeric part with increased stiffness effectively reduces the strain on the transistor and protects it from failure during normal operation. This method can effectively solve the problems of large chemical differences between the rigid island and the elastic substrate and the complexity of preparation, and can be well integrated into the existing preparation process. Transistors with a density of 340 cm^{-2} were integrated on the rigid island, and the transistor performance under the protection of the rigid island remained stable, changing no more than 5% when 100% strain was applied to the elastomer.

5. Research Progress of Stretchable System

Currently, stretchable circuits have made significant progress in conductivity, stretchability, and stress-resistant resistance. These features not only ensure the stability of the circuit under tensile strain but also demonstrate great potential in subsequent system integration.

Stretchable circuits have been extensively studied in various fields such as stretchable displays,[91,92] health monitoring[93,94] and human-computer interaction interfaces,[95] and have shown promising results. In this section, we will mainly focus on introducing flexible printed circuit boards and wearable health monitoring applications.

5.1. Elastic Printed Circuit Board

One novel concept proposed by Jiheong Kang and colleagues involves preparing a liquid metal network through the use of sound waves (**Figure 7**A). In this technique, the liquid metal inside an elastomer substrate forms a particle network of different sizes, with micron-sized particles forming the main frame and nano-sized particles encircling them.[22] This network maintains high stretchability and conductivity under strain, as the micron-sized particles deform from spherical to ellipsoidal while the nano-sized particles remain unchanged. Based on this percolation structure and deformation mechanism, a stretchable LED array and an electronic skin system with 38 chips were integrated and exhibited stable electrical properties under biaxial tensile deformation.

Traditionally, elastomers used in stretchable circuit systems are impermeable, which can reduce the comfort of users. In addition, the presence of substances such as sweat during health detection influenced the rapid, sensitive, and accurate detection of physiological signals. In response, Zijian Zheng and colleagues developed a hyperelastic conductor with high permeability, called the liquid metal fiber mat. This mat consists of a layer of liquid metal printed on an electrospun fiber film, which penetrates into the fiber film during the mechanical activation process.[96] The stretchable conductive fibers thus obtained exhibit good electrical conductivity and biocompatibility at 1800% omnidirectional tensile strain, making them suitable as electrodes for stretchable circuits. LEDs integrated into liquid metal fiber mats were encapsulated through electrospun fiber films and worked constantly under 500% tensile strain, 720° twisted strain, and even underwater without any liquid metal leakage, as shown in Figure 7B.

However, most stretchable circuits are currently designed on a plane, and in order to increase their integration, they are usually superimposed in the longitudinal direction.[97] This method may affect the thickness and sensitivity of the circuit, reducing its conformal contact with complex 3D planes and sensitivity and accuracy during use.[98] To address this issue, Nanjia Zhou and colleagues proposed a method for preparing a hydrogel stretchable circuit with a 3D structure (Figure 7C). This technique involves combining a curable hydrogel and stretchable silver hydrogel ink with 3D printing technology.[99] The hydrogel, acting as a framework, adopts an orthogonal crosslinking mechanism alginate-polyacrylamide double-network hydrogel. Electronic components such as LEDs and radio frequency identification chips are integrated into the hydrogels, which enable wirelessly powered LED coils without external metal lines. The LED continues to glow even under tensile conditions, making this a promising approach for developing stretchable circuits with improved sensitivity and accuracy.

5.2. Wearable Health Monitoring

In certain areas of human health detection applications, it is necessary to implant electronic components into the human body,[23] which can lead to additional risks and costs for patients during subsequent removal or replacement procedures. Additionally, health detection equipment often requires bulky and complex external equipment, which limits the practical application of real-time monitoring devices. To address these issues, biodegradable materials have been considered as potential materials for use in devices implanted in the human body.

John A. Rogers and his team proposed a health detection system that does not require an external power supply and utilizes biodegradable materials in the implanted par.[100] As shown in Figure 7D, the system consists of an implant module, a skin interface module, and an external control unit. A serpentine structure was chosen as the implanted part to prepare a biodegradable electrode for a temporary cardiac pacing system, which provides power to the radio frequency (RF) energy harvester in the body through wireless power supply, thus avoiding the need for complex wiring on the human body. The ultra-thin and stretchable design can reduce tissue interface stimulation as well as damage and can be customized to fit the geometry of individual patients.

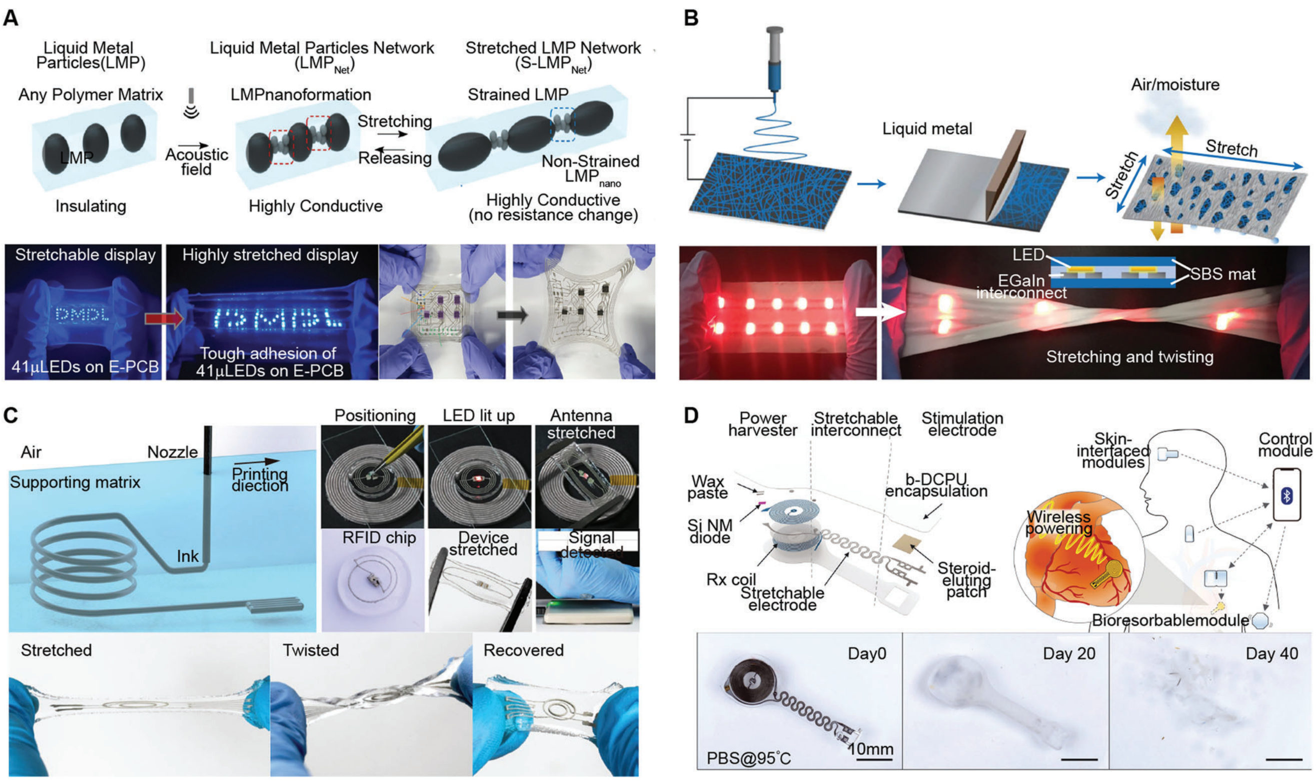

Figure 7. Research progress. A) A secondary structure network formed in the liquid metal particles to enhance the conductivity of the liquid metal during stretching. Reproduced with permission.[22] Copyright 2022, American Association for the Advancement of Science. B) Coating liquid metal on the fabric, embedding the liquid metal into the fabric by pre-stretching, can withstand 1100% strain without changing the resistance. Reproduced with permission.[96] Copyright 2021, Nature Publishing Group. C) 3D-printed stretchable 3D circuit. Reproduced with permission.[99] Copyright 2022, Nature Publishing Group. D) Serpentine electrodes prepared by biodegradable materials, which can be degraded while ensuring use, avoiding the risk of secondary surgery. Reproduced with permission.[100] Copyright 2022, American Association for the Advancement of Science.

The biodegradable material used in the body takes more than a month to decompose completely, which eliminates the need for reoperation.

6. Conclusion and Prospects

In summary, stretchable circuits offer several benefits, including normal operation during stretching, high performance, diversity, and electrical stability when integrated with other components. This makes them a promising area of development for various applications, particularly in health detection systems. Recent research and development of stretchable conductive materials, such as liquid metals, have significantly improved their conductivity, tensile strength, and robustness, allowing them to achieve stretchability that traditional metal materials cannot. However, these materials also present unique challenges, such as being incompatible with traditional PCB soldering processes and not withstanding high temperatures, which limit their potential use in certain applications. Nevertheless, continued research and development may lead to new solutions and applications in the future.

With the research and development of stretchable conductive materials, such as liquid metals, their conductivity, tensile strength, and robustness have been significantly improved. They can achieve stretchability that traditional metal materials cannot, and their electrical conductivity under stretching conditions is also higher than that of traditional metal materials. However, stretchable conductive materials have unique material properties, such as being in a liquid state, that make them incompatible with traditional PCB soldering processes. This presents a challenge when trying to establish stable connections with other components and materials. Additionally, stretchable conductive materials may not be able to withstand high temperatures, which limits their potential use in certain applications. Despite these challenges, continued research and development of stretchable conductive materials may lead to new solutions and applications in the future.

One promising approach is to design stretchable structures based on traditional metal conductive materials, which can be stably interconnected with other components using traditional welding technology. However, these structures have poor tensile properties, limiting their electrical stability under large strains. Additionally, the complex 2D/3D structure and the need for micro-nano processing techniques, such as photolithography, in the preparation process limit the integration degree of the circuit, resulting in complicated operation.

To improve the stability and functionality of stretchable circuits, researchers are exploring new methods, such as designing interface properties on the substrate to resolve differences in chemical properties between the conductive layer and the elastic substrate. This can effectively solve metal cracks caused by chemical differences under tensile conditions, increase the interface adhesion, and improve device firmness. Other methods including placing non-stretchable components on rigid islands solve the problem of electrical failure of rigid components under tension, increasing the diversity of usage scenarios. However, these methods are only applicable to specific polymer materials, which may reduce wearer comfort and increase the potential for inflammation.

Despite recent progress, research on stretchable circuits still faces many challenges, particularly in terms of scalable circuits for system integration and applications. Currently, stretchable circuits can only meet simple system integration, such as assembling LEDs or integrating battery packs. Further research is needed to address power supply, data transmission, and scalable manufacturing. Power supply is required to operate stretchable circuits, and conventional rigid batteries limit their development. Therefore, cleaner and more environmentally friendly energy sources, such as self-powered and solar cells, need to be explored. Data transmission is also a challenge, with wireless transmission being a common method. However, it has limitations, such as shorter transmission distances, slower speeds, higher latency, and data loss. Researchers must find appropriate data transmission methods for different signals to guarantee stability and reliability. Finally, stretchable circuits require miniaturization and sophistication research. The preparation process is complex and difficult to scale up for industrialization, limiting their practical application.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

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[1] Y. J. Jo, J. Ok, S. Y. Kim, T.-i. Kim, *Adv. Mater. Technol.* **2022**, *7*, 2001273.
[2] S. J. Benight, C. Wang, J. B. H. Tok, Z. Bao, *Prog. Polym. Sci.* **2013**, *38*, 1961.
[3] W. Wu, W. C. Poh, J. Lv, S. Chen, D. Gao, F. Yu, H. Wang, H. Fang, H. Wang, P. S. Lee, *Adv. Energy Mater.* **2023**, *13*, 2204103.
[4] Y. Lee, H. Cho, H. Yoon, H. Kang, H. Yoo, H. Zhou, S. Jeong, G. H. Lee, G. Kim, G.-T. Go, J. Seo, T.-W. Lee, Y. Hong, Y. Yun, *Adv. Mater. Technol.* **2023**, 2201067.
[5] H. Zhang, H. Chen, J.-H. Lee, E. Kim, K.-Y. Chan, H. Venkatesan, X. Shen, J. Yang, J.-K. Kim, *ACS Nano* **2023**, *17*, 5921.
[6] L. Wang, J. Wang, C. Fan, T. Xu, X. Zhang, *Chem. Eng. J.* **2023**, *455*, 140609.
[7] M. Xie, K. Hisano, M. Zhu, T. Toyoshi, M. Pan, S. Okada, O. Tsutsumi, S. Kawamura, C. Bowen, *Adv. Mater. Technol.* **2019**, *4*, 1800626.
[8] S. Choi, S. I. Han, D. Kim, T. Hyeon, D.-H. Kim, *Chem. Soc. Rev.* **2019**, *48*, 1566.
[9] C. Larson, B. Peele, S. Li, S. Robinson, M. Totaro, L. Beccai, B. Mazzolai, R. Shepherd, *Science* **2016**, *351*, 1071.
[10] Z. Yan, Y. Liu, J. Xiong, B. Wang, L. Dai, M. Gao, T. Pan, W. Yang, Y. Lin, *Adv. Mater.* **2023**, *35*, 2210238.
[11] N. Qaiser, A. N. Damdam, S. M. Khan, S. Bunaiyan, M. M. Hussain, *Adv. Funct. Mater.* **2021**, *31*, 2007445.
[12] J. H. Koo, D. Son, *Nat. Electron.* **2023**, *6*, 107.
[13] Y.-S. Guan, F. Ershad, Z. Rao, Z. Ke, E. C. da Costa, Q. Xiang, Y. Lu, X. Wang, J. Mei, P. Vanderslice, C. Hochman-Mendez, C. Yu, *Nat. Electron.* **2022**, *5*, 881.
[14] D. G. Mackanic, T.-H. Chang, Z. Huang, Y. Cui, Z. Bao, *Chem. Soc. Rev.* **2020**, *49*, 4466.
[15] J. Kim, S. Khan, P. Wu, S. Park, H. Park, C. Yu, W. Kim, *Nano Energy* **2021**, *79*, 105419.
[16] P. Li, H. P. Anwar Ali, W. Cheng, J. Yang, B. C. K. Tee, *Adv. Mater. Technol.* **2020**, *5*, 1900856.
[17] Y. Lee, W. J. Song, J. Y. Sun, *Mater. Today Phys.* **2020**, *15*, 100258.
[18] P. A. Lopes, D. F. Fernandes, A. F. Silva, D. G. Marques, A. T. de Almeida, C. Majidi, M. Tavakoli, *ACS Appl. Mater. Interfaces* **2021**, *13*, 14552.
[19] Q. Hua, J. Sun, H. Liu, R. Bao, R. Yu, J. Zhai, C. Pan, Z. L. Wang, *Nat. Commun.* **2018**, *9*, 244.
[20] J. Kang, J. Mun, Y. Zheng, M. Koizumi, N. Matsuhisa, H.-C. Wu, S. Chen, J. B. H. Tok, G. H. Lee, L. Jin, Z. Bao, *Nat. Nanotechnol.* **2022**, *17*, 1265.
[21] W. Wang, S. Wang, R. Rastak, Y. Ochiai, S. Niu, Y. Jiang, P. K. Arunachala, Y. Zheng, J. Xu, N. Matsuhisa, X. Yan, S.-K. Kwon, M. Miyakawa, Z. Zhang, R. Ning, A. M. Foudeh, Y. Yun, C. Linder, J. B. H. Tok, Z. Bao, *Nat. Electron.* **2021**, *4*, 143.
[22] W. Lee, H. Kim, I. Kang, H. Park, J. Jung, H. Lee, H. Park, J. S. Park, J. M. Yuk, S. Ryu, J.-W. Jeong, J. Kang, *Science* **2022**, *378*, 637.
[23] H. Hu, H. Huang, M. Li, X. Gao, L. Yin, R. Qi, R. S. Wu, X. Chen, Y. Ma, K. Shi, C. Li, T. M. Maus, B. Huang, C. Lu, M. Lin, S. Zhou, Z. Lou, Y. Gu, Y. Chen, Y. Lei, X. Wang, R. Wang, W. Yue, X. Yang, Y. Bian, J. Mu, G. Park, S. Xiang, S. Cai, P. W. Corey, et al., *Nature* **2023**, *613*, 667.
[24] C. W. Park, Y. G. Moon, H. Seong, S. W. Jung, J.-Y. Oh, B. S. Na, N.-M. Park, S. S. Lee, S. G. Im, J. B. Koo, *ACS Appl. Mater. Interfaces* **2016**, *8*, 15459.
[25] D. G.Marques, P. A.Lopes, A. T. de Almeida, C. Majidi, M. Tavakoli, *Lab Chip* **2019**, *19*, 897.
[26] S. Chen, S. Fan, J. Qi, Z. Xiong, Z. Qiao, Z. Wu, J. C. Yeo, C. T. Lim, *Adv. Mater.* **2023**, *35*, 2208569.
[27] K. B. Ozutemiz, J. Wissman, O. B. Ozdoganlar, C. Majidi, *Adv. Mater. Interfaces* **2018**, *5*, 1701596.
[28] J.-E. Park, H. S. Kang, M. Koo, C. Park, *Adv. Mater.* **2020**, *32*, 2002178.
[29] M. A. H. Khondoker, A. Ostashek, D. Sameoto, *Adv. Eng. Mater.* **2019**, *21*, 1900060.
[30] J. Zhang, B. Ma, G. Chen, Y. Chen, C. Xu, Q. Hao, C. Zhao, H. Liu, *ACS Appl. Mater. Interfaces* **2022**, *14*, 53405.
[31] M. Tavakoli, M. H. Malakooti, H. Paisana, Y. Ohm, D. G.Marques, P. A. Lopes, A. P. Piedade, A. T. de Almeida, C. Majidi, *Adv. Mater.* **2018**, *30*, 1801852.

[32] S. Wang, Y. Nie, H. Zhu, Y. Xu, S. Cao, J. Zhang, Y. Li, J. Wang, X. Ning, D. Kong, *Sci. Adv.* **2022**, *8*, eabl5511.
[33] G. Li, X. Wu, D.-W. Lee, *Sens. Actuators, B* **2015**, *221*, 1114.
[34] M.-g. Kim, D. K. Brown, O. Brand, *Nat. Commun.* **2020**, *11*, 1002.
[35] B. A. Gozen, A. Tabatabai, O. B. Ozdoganlar, C. Majidi, *Adv. Mater.* **2014**, *26*, 5211.
[36] A. F. Silva, H. Paisana, T. Fernandes, J. Góis, A. Serra, J. F. J. Coelho, A. T. de Almeida, C. Majidi, M. Tavakoli, *Adv. Mater. Technol.* **2020**, *5*, 2000343.
[37] M. Kim, H. Lim, S. H. Ko, *Adv. Sci.* **2023**, *10*, 2205795.
[38] P. Wu, J. Fu, Y. Xu, Y. He, *ACS Appl. Mater. Interfaces* **2022**, *14*, 13458.
[39] J. Dong, Y. Peng, L. Pu, K. Chang, L. Li, C. Zhang, P. Ma, Y. Huang, T. Liu, *Nano Lett.*. **2022**, *22*, 7597.
[40] X. Zheng, W. Cao, X. Hong, L. Zou, Z. Liu, P. Wang, C. Li, *Small* **2023**, *19*, 2208134.
[41] J. Shi, S. Liu, L. Zhang, B. Yang, L. Shu, Y. Yang, M. Ren, Y. Wang, J. Chen, W. Chen, Y. Chai, X. Tao, *Adv. Mater.* **2020**, *32*, 1901958.
[42] S. Zhu, M. Wang, Z. Qiang, J. Song, Y. Wang, Y. Fan, Z. You, Y. Liao, M. Zhu, C. Ye, *Chem. Eng. J.* **2021**, *406*, 127140.
[43] Z. Ouyang, D. Xu, H.-Y. Yu, S. Li, Y. Song, K. C. Tam, *Chem. Eng. J.* **2022**, *428*, 131289.
[44] Y. Luo, Y. Li, P. Sharma, W. Shou, K. Wu, M. Foshey, B. Li, T. Palacios, A. Torralba, W. Matusik, *Nat. Electron.* **2021**, *4*, 314.
[45] M. Park, J. Im, M. Shin, Y. Min, J. Park, H. Cho, S. Park, M.-B. Shim, S. Jeon, D.-Y. Chung, J. Bae, J. Park, U. Jeong, K. Kim, *Nat. Nanotechnol.* **2012**, *7*, 803.
[46] X.-X. Wang, G.-F. Yu, J. Zhang, M. Yu, S. Ramakrishna, Y.-Z. Long, *Prog. Mater. Sci.* **2021**, *115*, 100704.
[47] A. S. Levitt, M. Alhabeb, C. B. Hatter, A. Sarycheva, G. Dion, Y. Gogotsi, *J. Mater. Chem. A* **2019**, *7*, 269.
[48] B. Bessaire, M. Mathieu, V. Salles, T. Yeghoyan, C. Celle, J.-P. Simonato, A. Brioude, *ACS Appl. Mater. Interfaces* **2017**, *9*, 950.
[49] Y. Yang, T. Vervust, S. Dunphy, S. Van Put, B. Vandecasteele, K. Dhaenens, L. Degrendele, L. Mader, L. De Vriese, T. Martens, M. Kaufmann, T. Sekitani, J. Vanfleteren, *Adv. Electron. Mater.* **2018**, *4*, 1800071.
[50] R. Lin, H.-J. Kim, S. Achavananthadith, Z. Xiong, J. K. W. Lee, Y. L. Kong, J. S. Ho, *Nat. Commun.* **2022**, *13*, 2190.
[51] D. Shukla, Y. Liu, Y. Zhu, *Nanoscale* **2023**, *15*, 2767.
[52] W. Zu, Y. Ohm, M. R. Carneiro, M. Vinciguerra, M. Tavakoli, C. Majidi, *Adv. Mater. Technol.* **2022**, *7*, 2200534.
[53] U. Kraft, F. Molina-Lopez, D. Son, Z. Bao, B. Murmann, *Adv. Electron. Mater.* **2020**, *6*, 1900681.
[54] Y. Oh, I. S. Yoon, C. Lee, S. H. Kim, B.-K. Ju, J.-M. Hong, *J. Mater. Chem. C* **2017**, *5*, 11733.
[55] Y. Kang, G. Wang, S. Zhao, J. Li, L. Di, Y. Feng, J. Yin, J. Zhu, *Small* **2020**, *16*, 2004793.
[56] J. Wang, G. Cai, S. Li, D. Gao, J. Xiong, P. S. Lee, *Adv. Mater.* **2018**, *30*, 1706157.
[57] Y. Meng, T. Ma, F. J. Pavinatto, J. D. MacKenzie, *ACS Appl. Mater. Interfaces* **2019**, *11*, 9190.
[58] N. Matsuhisa, M. Kaltenbrunner, T. Yokota, H. Jinno, K. Kuribara, T. Sekitani, T. Someya, *Nat. Commun.* **2015**, *6*, 7461.
[59] H. Jin, M. O. G. Nayeem, S. Lee, N. Matsuhisa, D. Inoue, T. Yokota, D. Hashizume, T. Someya, *ACS Nano* **2019**, *13*, 7905.
[60] N. Matsuhisa, D. Inoue, P. Zalar, H. Jin, Y. Matsuba, A. Itoh, T. Yokota, D. Hashizume, T. Someya, *Nat. Mater.* **2017**, *16*, 834.
[61] J. Park, J. S. Myung, D. Cho, T. Kim, S. Y. Lee, Y. Kim, Y. Choi, S. Jung, *Adv. Electron. Mater.* **2023**, *9*, 2201021.
[62] T. Wang, Q. Liu, H. Liu, B. Xu, H. Xu, *Adv. Mater.* **2022**, *34*, 2202418.
[63] M. Hu, X. Cai, Q. Guo, B. Bian, T. Zhang, J. Yang, *ACS Nano* **2016**, *10*, 396.
[64] J. Yuan, Y. Zhang, G. Li, S. Liu, R. Zhu, *Adv. Funct. Mater.* **2022**, *32*, 2204878.
[65] S. H. Kim, H. Seo, J. Kang, J. Hong, D. Seong, H.-J. Kim, J. Kim, J. Mun, I. Youn, J. Kim, Y.-C. Kim, H.-K. Seok, C. Lee, J. B. H. Tok, Z. Bao, D. Son, *ACS Nano* **2019**, *13*, 6531.
[66] C. M. Ajmal, S. Cha, W. Kim, K. P. Faseela, H. Yang, S. Baik, *Sci. Adv.* **2022**, *8*, eabn3365.
[67] Y. Lee, B. J. Kim, L. Hu, J. Hong, J.-H. Ahn, *Mater. Today* **2022**, *53*, 51.
[68] X. Chen, Y. Li, X. Wang, H. Yu, *ACS Appl. Mater. Interfaces* **2022**, *14*, 36227.
[69] T. C. Shyu, P. F. Damasceno, P. M. Dodd, A. Lamoureux, L. Xu, M. Shlian, M. Shtein, S. C. Glotzer, N. A. Kotov, *Nat. Mater.* **2015**, *14*, 785.
[70] H. Li, Z. Wang, M. Sun, H. Zhu, H. Liu, C. Y. Tang, L. Xu, *Adv. Funct. Mater.* **2022**, *32*, 2202792.
[71] C. Kang, S.-W. Kim, W. Kim, D. Choi, H.-K. Kim, *Adv. Mater. Interfaces* **2023**, *10*, 2202477.
[72] S. Xu, Z. Yan, K.-I. Jang, W. Huang, H. Fu, J. Kim, Z. Wei, M. Flavin, J. McCracken, R. Wang, A. Badea, Y. Liu, D. Xiao, G. Zhou, J. Lee, H. U. Chung, H. Cheng, W. Ren, A. Banks, X. Li, U. Paik, R. G. Nuzzo, Y. Huang, Y. Zhang, J. A. Rogers, *Science* **2015**, *347*, 154.
[73] K.-I. Jang, K. Li, H. U. Chung, S. Xu, H. N. Jung, Y. Yang, J. W. Kwak, H. H. Jung, J. Song, C. Yang, A. Wang, Z. Liu, J. Y. Lee, B. H. Kim, J.-H. Kim, J. Lee, Y. Yu, B. J. Kim, H. Jang, K. J. Yu, J. Kim, J. W. Lee, J.-W. Jeong, Y. M. Song, Y. Huang, Y. Zhang, J. A. Rogers, *Nat. Commun.* **2017**, *8*, 15894.
[74] D. Wang, L. Dong, G. Gu, *Adv. Funct. Mater.* **2023**, *33*, 2208849.
[75] X. Ma, X. Wu, S. Cao, Y. Zhao, Y. Lin, Y. Xu, X. Ning, D. Kong, *Adv. Sci.* **2023**, *10*, 2205343.
[76] Q. Mu, L. Wang, C. K. Dunn, X. Kuang, F. Duan, Z. Zhang, H. J. Qi, T. Wang, *Addit. Manuf.* **2017**, *18*, 74.
[77] Y. Gao, F. Guo, P. Cao, J. Liu, D. Li, J. Wu, N. Wang, Y. Su, Y. Zhao, *ACS Nano* **2020**, *14*, 3442.
[78] S. Xu, Y. Zhang, L. Jia, K. E. Mathewson, K.-I. Jang, J. Kim, H. Fu, X. Huang, P. Chava, R. Wang, S. Bhole, L. Wang, Y. J. Na, Y. Guan, M. Flavin, Z. Han, Y. Huang, J. A. Rogers, *Science* **2014**, *344*, 70.
[79] Z. Huang, Y. Hao, Y. Li, H. Hu, C. Wang, A. Nomoto, T. Pan, Y. Gu, Y. Chen, T. Zhang, W. Li, Y. Lei, N. Kim, C. Wang, L. Zhang, J. W. Ward, A. Maralani, X. Li, M. F. Durstock, A. Pisano, Y. Lin, S. Xu, *Nat. Electron.* **2018**, *1*, 473.
[80] S. Jang, C. Kim, J. J. Park, M. L. Jin, S. J. Kim, O. O. Park, T.-S. Kim, H.-T. Jung, *Small* **2018**, *14*, 1702818.
[81] M. Kong, I. You, G. Lee, G. Park, J. Kim, D. Park, U. Jeong, *Adv. Mater.* **2021**, *33*, 2100299.
[82] Q. Zhang, S. Niu, L. Wang, J. Lopez, S. Chen, Y. Cai, R. Du, Y. Liu, J.-C. Lai, L. Liu, C.-H. Li, X. Yan, C. Liu, J. B. H. Tok, X. Jia, Z. Bao, *Adv. Mater.* **2018**, *30*, 1801435.
[83] P. A. Lopes, B. C. Santos, A. T. de Almeida, M. Tavakoli, *Nat. Commun.* **2021**, *12*, 4666.
[84] N. Naserifar, P. R. LeDuc, G. K. Fedder, *Adv. Mater.* **2016**, *28*, 3584.
[85] M. Cai, S. Nie, Y. Du, C. Wang, J. Song, *ACS Appl. Mater. Interfaces* **2019**, *11*, 14340.
[86] H. Li, Y. Xu, X. Li, Y. Chen, Y. Jiang, C. Zhang, B. Lu, J. Wang, Y. Ma, Y. Chen, Y. Huang, M. Ding, H. Su, G. Song, Y. Luo, X. Feng, *Adv. Healthcare Mater.* **2017**, *6*, 1601013.
[87] Y. Cao, G. Zhang, Y. Zhang, M. Yue, Y. Chen, S. Cai, T. Xie, X. Feng, *Adv. Funct. Mater.* **2018**, *28*, 1804604.
[88] T. Sekitani, U. Zschieschang, H. Klauk, T. Someya, *Nat. Mater.* **2010**, *9*, 1015.
[89] J. C. Yang, S. Lee, B. S. Ma, J. Kim, M. Song, S. Y. Kim, D. W. Kim, T.-S. Kim, S. Park, *Sci. Adv.* **2022**, *8*, eabn3863.
[90] S. Wang, J. Xu, W. Wang, G.-J. N. Wang, R. Rastak, F. Molina-Lopez, J. W. Chung, S. Niu, V. R. Feig, J. Lopez, T. Lei, S.-K. Kwon, Y. Kim, A. M. Foudeh, A. Ehrlich, A. Gasperini, Y. Yun, B. Murmann, J. B. H. Tok, Z. Bao, *Nature* **2018**, *555*, 83.

[91] Y.-Q. Zheng, Y. Liu, D. Zhong, S. Nikzad, S. Liu, Z. Yu, D. Liu, H.-C. Wu, C. Zhu, J. Li, H. Tran, J. B. H. Tok, Z. Bao, *Science* **2021**, *373*, 88.
[92] Y. Kim, J. M. Suh, J. Shin, Y. Liu, H. Yeon, K. Qiao, H. S. Kum, C. Kim, H. E. Lee, C. Choi, H. Kim, D. Lee, J. Lee, J.-H. Kang, B.-I. Park, S. Kang, J. Kim, S. Kim, J. A. Perozek, K. Wang, Y. Park, K. Kishen, L. Kong, T. Palacios, J. Park, M.-C. Park, H.-j. Kim, Y. S. Lee, K. Lee, S.-H. Bae, et al., *Science* **2022**, *377*, 859.
[93] Y. Xu, Y. Su, X. Xu, B. Arends, G. Zhao, D. N. Ackerman, H. Huang, S. P. Reid, J. L. Santarpia, C. Kim, Z. Chen, S. Mahmoud, Y. Ling, A. Brown, Q. Chen, G. Huang, J. Xie, Z. Yan, *Sci. Adv.* **2023**, *9*, eadf0575.
[94] J. Gu, F. Li, Y. Zhu, D. Li, X. Liu, B. Wu, H.-A. Wu, X. Fan, X. Ji, Y. Chen, J. Liang, *Adv. Mater.* **2023**, *35*, 2209527.
[95] S. Han, K. Kim, S. Y. Lee, S. Moon, J.-Y. Lee, *Adv. Mater.* **2023**, *35*, 2210112.
[96] Z. Ma, Q. Huang, Q. Xu, Q. Zhuang, X. Zhao, Y. Yang, H. Qiu, Z. Yang, C. Wang, Y. Chai, Z. Zheng, *Nat. Mater.* **2021**, *20*, 859.
[97] H. Song, G. Luo, Z. Ji, R. Bo, Z. Xue, D. Yan, F. Zhang, K. Bai, J. Liu, X. Cheng, W. Pang, Z. Shen, Y. Zhang, *Sci. Adv.* **2022**, *8*, eabm3785.
[98] G. Li, M. Zhang, S. Liu, M. Yuan, J. Wu, M. Yu, L. Teng, Z. Xu, J. Guo, G. Li, Z. Liu, X. Ma, *Nat. Electron.* **2023**, *6*, 154.
[99] Y. Hui, Y. Yao, Q. Qian, J. Luo, H. Chen, Z. Qiao, Y. Yu, L. Tao, N. Zhou, *Nat. Electron.* **2022**, *5*, 893.
[100] Y. S. Choi, H. Jeong, R. T. Yin, R. Avila, A. Pfenniger, J. Yoo, J. Y. Lee, A. Tzavelis, Y. J. Lee, S. W. Chen, H. S. Knight, S. Kim, H.-Y. Ahn, G. Wickerson, A. Vázquez-Guardado, E. Higbee-Dempsey, B. A. Russo, M. A. Napolitano, T. J. Holleran, L. A. Razzak, A. N. Miniovich, G. Lee, B. Geist, B. Kim, S. Han, J. A. Brennan, K. Aras, S. S. Kwak, J. Kim, E. A. Waters, et al., *Science* **2022**, *376*, 1006.

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