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*Review*

# E-Polymers: Applications in Biological Interfaces and Organisms

**Weixin Dou [1], Sihong Wang [2,*] and Rusen Yang [1,*]**

[1] School of Advanced Materials and Nanotechnology, Xidian University, Xi'an 710126, China; 15501835079@163.com
[2] Pritzker School of Molecular Engineering, The University of Chicago, Chicago, IL 60637, USA
* Correspondence: sihongwang@uchicago.edu (S.W.); rsyang@xidian.edu.cn (R.Y.)

**Abstract:** Future electronics will play a more critical role in people's lives, as reflected in the realization of advanced human–machine interfaces, disease detection, medical treatment, and health monitoring. The current electronic products are rigid, non-degradable, and cannot repair themselves. Meanwhile, the human body is soft, dynamic, stretchable, degradable, and self-healing. Consequently, it is valuable to develop new electronic materials with skin-like properties that include stretchability, inhibition of invasive reactions, self-healing, long-term durability, and biodegradability. These demands have driven the development of a new generation of electronic materials with high-electrical performance and skin-like properties, among which e-polymers are increasingly being more extensively investigated. This review focuses on recent advances in synthesizing e-polymers and their applications in biointerfaces and organisms. Discussions include the synthesis and properties of e-polymers, the interrelationships between engineered material structures and human interfaces, and the application of implantable and wearable systems for sensors and energy harvesters. The final section summarizes the challenges and future opportunities in the evolving materials and biomedical research field.

**Keywords:** e-polymers; stretchability; bio-interface; 3D structure; sensor; energy harvesting; bio-integrated

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## 1. Introduction

In the era of the Internet of Things, electronic products focus on connecting more and more objects to effectively collect and exchange information, ultimately creating a universal internet [1,2]. Collecting data from the human body is the core of developing this idea, potentially leading to intelligence and universal healthcare [3,4]. However, this will require electronic devices that can interface with various parts of the human body and collect signals with sufficient quality, resolution, and stability [5,6]. In the foreseeable time, electronic products are expected to play an even more critical role in healthcare, medicine, biological research, and human–machine interfaces [7–9]. The compatible and tight integration of electronic devices with the human body can be achieved by improving biocompatibility [10], enhancing compatibility, and inhibiting intrusive reactions [11,12]. There is an urgent need for research on a new generation of electronic materials [13–15] that are stretchable, self-healing, and biodegradable. They also have similar skin properties and high electronic properties [16–18]. They have many applications in wearable electronics, electronic prosthetic skin [19–21], and implanted biomedical electronics [22].

E-polymers, also known as conducting polymers, are a class of materials that exhibit both electrical conductivity and the mechanical properties of polymers [23]. The use of e-polymer materials in daily life is becoming increasingly widespread, especially in the field of biology [24,25]. Since the manufacturing cost of e-polymer implants is relatively low and e-polymers also react, causing different chemical molecules to attach to the surface of the

implant, they are more compatible with the surrounding environment of the body [26,27]. Some e-polymers are biodegradable in the body. If used for temporary implants, the advantage of these polymers is that they can gradually degrade in the body after performing their functions [28–30], thereby reducing the possibility of any long-term complications [31]. Polymers and their composite materials can be designed to have inherent tensile properties while maintaining their high performance, making them favorable candidates for the next generation of skin-inspired electronic materials [32,33]. In addition, combining self-healing polymers and biodegradable materials can improve the durability and biocompatibility of electronic skin [34,35]. Wearable electronic devices such as smartwatches and fitness trackers have sensors that monitor various physical activities, including heart rate, temperature, blood pressure, and exercise [36,37]. Integrating flexible and stretchable electronic materials can improve the comfort and wearability of these devices. In addition, self-healing e-polymers can prevent damage caused by continuous bending and stretching, while biodegradable materials can ensure the ecological friendliness of the product [38,39]. Strain engineering is an effective strategy for introducing mechanical rigid electronic materials into external tensile properties. In any soft interface, whether from human skin or soft robots, forces generated through physical contact in the body or within the body are the primary means of sensing haptic interaction [40,41]. Interestingly, soft pressure sensors can convert mechanical stimuli into electrical signals due to their pressure properties. They can be used in medical implants, wearable health monitoring, electronic prosthetic skin, human-computer interaction, and artificial intelligence. The matching of electronic devices with application objectives is presented using flexible and stretchable materials and utilizing 3D geometric shapes [42,43], micro-pattern designs, grids, and cavity structural designs. This emerging design strategy for stretchable devices overcomes the shortcomings of traditional instruments, such as rigidity, volume, and rigid shape factors [44–46].

This review highlights the latest developments in the emerging "e-polymer-based materials for biological applications" field. We first considered the working mechanism and material composition of e-polymers and then outlined the latest developments based on e-polymer materials. In contrast, this review investigates the progress in the application of e-polymer-based materials in biology, linking the development of e-polymer-based materials with physiological signal patterns that may have widespread applications in human health. The first section provides an overview of recent developments in e-polymers and their synthesis based on e-polymer materials. In an attempt to comprehend the mechanism of synthesis at the molecular level, we first introduce the methods for the synthesis of e-polymer materials. In the second section, we discuss in detail the semiconducting, sensing, and mechanical properties of e-polymer materials. In the third section, we also summarise the potential applications of e-polymer materials in energy harvesting, sensors, and other areas, focusing on the most advanced types of sensors. Finally, we outline the remaining critical challenges and developments and outline the opportunities for sustainable development based on e-polymer materials in the biological field.

## 2. Synthesis and Design of E-Polymers

In our daily lives, we encounter a wide variety of e-polymers, some of which are natural and some of which are synthetic [47]. Sufficient flexibility and biocompatibility compared to inorganic materials, as well as a range of electron transport, chemical functionality, and tailored mechanical and optical properties, are all advantages that make e-polymers very attractive [48]. E-polymers utilize specialized chemical substances and composite materials for biological applications [49,50]. The synthetic-based electronic polymers encompass conducting hydrogels and ionogels, electrochemical transistors, and topological supramolecular networks. The synthesis of e-polymers involves the polymerization of monomers with conjugated structures to form long-chain polymers that exhibit electronic and optoelectronic properties.

E-polymers based on artificial synthesis are those that are synthesized from monomers or building blocks that are not found in nature. Unlike polymers based on biomolecules,

which use natural biopolymers as precursors, these polymers are designed and synthesized using synthetic chemistry techniques [51,52]. One approach to synthesizing polymers is through step-growth polymerization, which involves gradually forming a polymer chain through reacting reactive groups at the ends of the growing chain with functional groups on monomers [53,54]. This technique can create various polymers with different properties, such as polycarbonates, polyesters, and polyamides. Another approach is through chain-growth polymerization, which involves initiating a polymerization reaction at a reactive site on a monomer, followed by adding more monomers to the growing chain [55,56]. In addition to these traditional polymerization methods, researchers are also exploring new strategies for the synthesis of polymers, such as click chemistry, which involves the selective reaction of two functional groups to form a covalent bond. Click reactions can be used to create complex polymer structures with precise control over their size and shape [55,57,58]. Polymer semiconductors have shown unique development advantages in the development of human-integrated electronic products due to their solution processability and mechanical flexibility. However, many of the functional characteristics required in this application field are transferred to conjugated polymers, which are combined with effective charge transfer properties. In a study, Li et al. developed a "click-to-polymer" (CLIP) synthesis strategy that utilizes click reactions to attach different types of functional units to pre-synthesized conjugated polymer precursors (Figure 1A) [59]. It has been proven that the functionalized polymer of the method can still maintain good carrier mobility. The functional properties of conjugated polymers can be greatly enriched by using this synthetic method.

The most commonly used materials for conventional planar electronic devices are inorganic, but the brittle and mechanical properties of inorganic materials are unsuitable for applications in the biological field [60]. Extensive research has attempted to find alternative materials that bypass mechanical limitations without sacrificing functionality or performance. Materials ranging from single-crystal silicon nanofilms, nanowires, and nanobelts to conjugated small-molecule organic polycrystalline films are semiconductor component choices for such thin flexible devices and are valuable for the research and development of flexible devices [61]. Scalable bioelectronic devices are based on flexible and conductive organic materials that allow rational interfaces for biocompatible integration with the human body. In a study, Jiang et al. developed a molecular engineering strategy based on topological supramolecular networks that decouple competitive effects from multiple molecular components (Figure 1B) [62]. Under physiological conditions, both high conductivity and crack initiation strain were obtained, exhibiting direct photosensitivity to the cell scale. Further, the stable EMG signals of the octopus were collected, local neuromodulation was conducted, and the specific activities of the organ were conducted through the exemplary brainstem controller.

The basic principle of complementary metal-oxide-semiconductor technology is to utilize the complementary properties of p-type and n-type metal-oxide-semiconductor materials to achieve an efficient operation of circuits [63,64]. For the construction of CMOS logic circuits and p–n junction devices, n-type semiconductors play a crucial role [65]. Among various electronic deficient components, cyanide-functionalized hydrocarbons are emerging to achieve high-performance n-type organic and polymer semiconductors [31]. In a study, Li et al. developed a large number of n-type organic semiconductors and polymer semiconductors based on these cyanide functional compositions, which show many suppressed frontier molecular orbitals (FMOs) compared to their non-cyanide analogs (Figure 1C) [66]. The incorporation of cyanide significantly inhibits FMOs in semiconductors, leading to an n-type transport. A series of new electron-deficient components can be generated to build n-type organic and polymer semiconductors, which ultimately manifests itself in enhanced device performance. For further development, integrated design strategies are a feasible way to achieve these goals, thereby promoting the construction of high-performance n-type semiconductors. The synthesis of e-polymers based on artificial synthesis offers a powerful tool for creating new materials with tailored properties and functions, including applications in materials science, electronics, and biomedicine. In a

study, Wang et al. presented a fundamentally stretchable polymer transistor array with an unprecedented device density of 347 transistors per square centimeter (Figure 1D) [67]. Consequently, transistor arrays essentially constitute stretchable skin electronics, including active matrices for sensing arrays as well as analog and digital circuit elements.

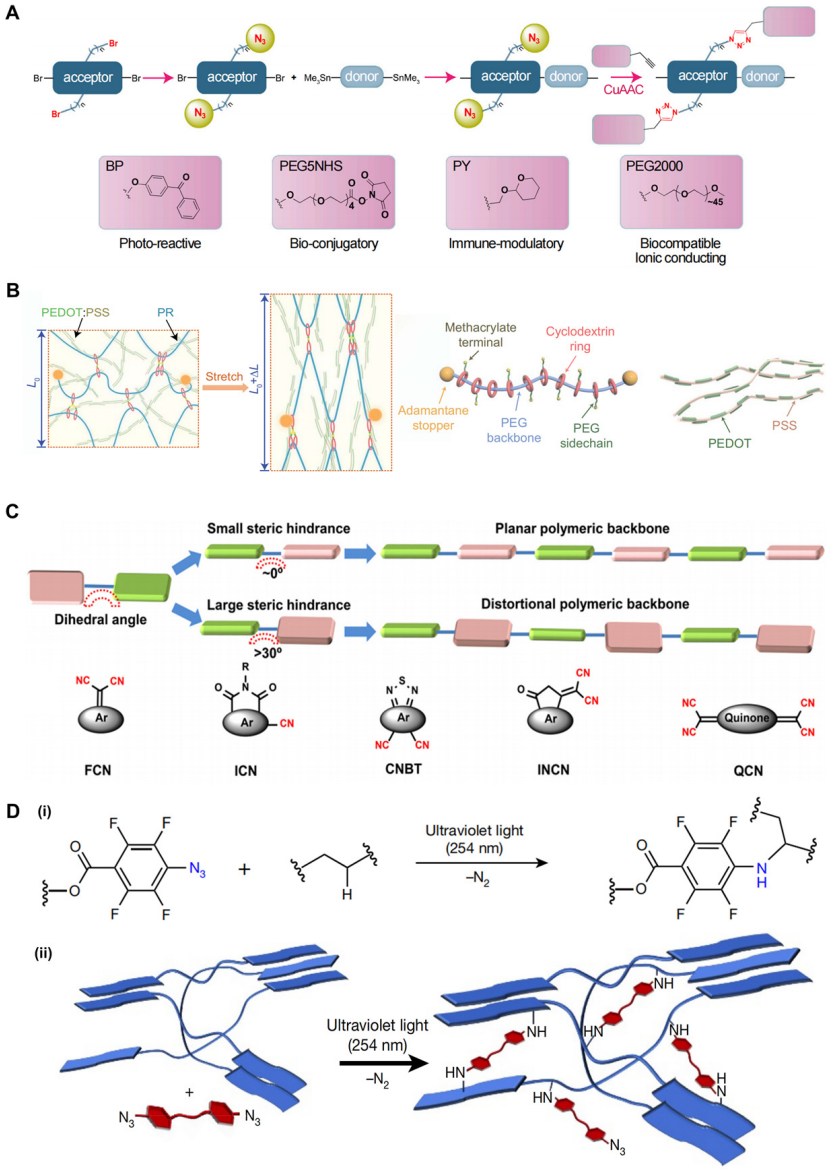


**Figure 1.** (**A**) A general and simple strategy for CLIP synthesis has been established. This strategy employs a one-step multifunctional reaction, which results in the covalent connection of various functional units. Adapted from Ref. [59] with permission. Copyright 2021 Elsevier B.V. (**B**) Schematic representation of an intrinsically stretchable topological supramolecular network with key molecular building blocks of PR monomers. Adapted from Ref. [62] with permission. Copyright 2022 American Association for the Advancement of Science. (**C**) Schematic representation of the planar backbone and the five cyano-functional group structural units in conjugated polymers. Adapted from Ref. [66] with permission. Copyright 2023 Royal Society of Chemistry. (**D**) The azide crosslinking reaction, which is initiated by UV light, is based on the reaction between the azide and CH groups. (i) An azide-crosslinking reaction, which is initiated by ultraviolet light and is based on the reaction between azide groups and CH groups. (ii) How the polymer-chain network in an elastomer (blue, with rectangular planes representing rigid segments and tortuous lines representing soft segments) becomes crosslinked by azides (red) into a three-dimensional network. Adapted from Ref. [67] with permission. Copyright 2018 Springer Nature.

## 3. Properties of E-Polymers

E-polymers have many properties and unique electrical and photovoltaic properties that allow them to be used in a variety of applications [67–69]. Many e-polymers have high strength and durability, making them ideal for use in products that require durability and robustness. In addition, inherently stretchable semiconductor polymers use molecular structure engineering, such as length and branching of alkyl side chains, molecular weight, and the design of blends containing both rigid and flexible electronic blocks, to make the copolymers stretchable [70,71]. E-polymers can conduct electricity, which sets them apart from traditional insulating polymers. Depending on the chemical structure, doping, and processing conditions, e-polymers have different conductivity properties ranging from semiconducting behavior to metallic conductivity [72–74]. E-polymers can exhibit interesting optical properties, including absorption and emission of light in the visible and near-infrared regions. The energy band gap of an e-polymer can be tuned by changing its chemical structure, thereby controlling the wavelength of the emitted light. E-polymers can transport charge carriers (electrons or holes) through a conjugated backbone. The mobility of charge carriers in e-polymers is influenced by factors such as polymer crystallinity, chain organization, and molecular weight. Efficient charge transport is critical for applications such as organic solar cells, transistors, and conductive coatings. One of the advantages of e-polymers is their flexibility and processability [75–77]. They can be made into films, fibers, or coatings by a variety of techniques such as solution casting, spin-coating, printing, or vapor phase deposition. This flexibility allows e-polymers to be integrated into flexible and lightweight devices, opening up possibilities for wearable electronics and flexible displays. E-polymers can undergo redox reactions, meaning they can be oxidized or reduced while maintaining a conjugated structure [78]. Compared to traditional inorganic semiconductors, e-polymers typically have good environmental stability. However, their stability can vary depending on factors such as polymer selection, device design, and operating conditions [79]. In addition, e-polymers typically have potential environmental advantages over conventional inorganic electronic materials. They can be synthesized from abundant renewable resources, and some polymers are biocompatible [80]. Beneficially, e-polymer devices have the potential for low-cost manufacturing processes, thereby reducing the environmental impact of the manufacturing process. These properties of e-polymers make them attractive for a variety of applications, including organic electronics, optoelectronics, sensors, energy conversion and storage, smart textiles, and biomedical devices [81].

A growing and widespread concern is the application of e-polymers to bio-interfaces and organisms [82,83]. There is an imperative demand to synthesize novel and sustainable e-polymers for bio-interface and organism applications that can functionally replace the existing e-polymers or exhibit their properties and advantages. In a study, Kang et al. described a new class of polymeric material crosslinked through rationally designed multistrength hydrogen-bonding interactions (Figure 2A) [84]. A supramolecular polymer film constructed through a mixture of strong and weak crosslinking hydrogen bonds is described. The resulting polymer possesses various mechanical properties required for electronic skin applications, such as stretchability, toughness, and the ability to autonomously self-heal even in water. As this polymer is easy to manipulate, capacitive strain-sensing electronic skins are designed and fabricated to be highly resilient and resistant to vandalism. The exhibits feature an advanced structure, excellent thermo–mechanical properties, higher stability, lower flammability, better processing conditions, and improved appearance. In a study, Li et al. demonstrated stretchable transistor arrays and active matrix circuits with moduli below 10 kPa (Figure 2B) [85]. Due to an improved adaptability to irregular and dynamic surfaces, an ultrasoft device fabricated using a soft sandwich design enables electrophysiological recording of the isolated heart. High adaptability, spatial stability, and minimal impact on ventricular pressure were achieved. Additionally, testing has demonstrated the benefits of inhibiting foreign body reactions for long-term implantation, resulting in superior in vivo biocompatibility. E-polymers have similar electrical and electrochemical properties to traditional semiconductors and metals, thus receiving

widespread attention in both basic and practical research [86,87]. The electrical conductivity of organic radical polymers is much higher than expected, and organic radical polymers have unusual electronic properties (Figure 2C) [88]. Conductivity can be improved in two approaches. On the one hand, this can be accomplished by the synthesis of molecular structures with a relatively large dispersion of π-bonds; the higher the dispersion, the improved conductivity of the conjugated structure. Consequently, improving the intrinsic conductivity of polymers from the perspective of the molecular structure is an optimal solution. In addition, improving production processes and preparing polymer materials with larger molecular weights and more regular structures are also important means to improve their conductivity. On the other hand, the chemical doping of conjugated structures is an effective way to enhance the conductivity of polymer materials by introducing anions (p-type doping) or cations (n-type doping) on the polymer chain through doping methods to reduce energy barriers and facilitate electron migration. The commonly used dopants include iodine, arsenic pentafluoride, antimony hexafluoride, silver perchlorate, etc. After the dopants are saturated, the conductivity of the material will not change [89,90]. Therefore, it will be important to find suitable doping agents and dope them reasonably with conductive polymers.

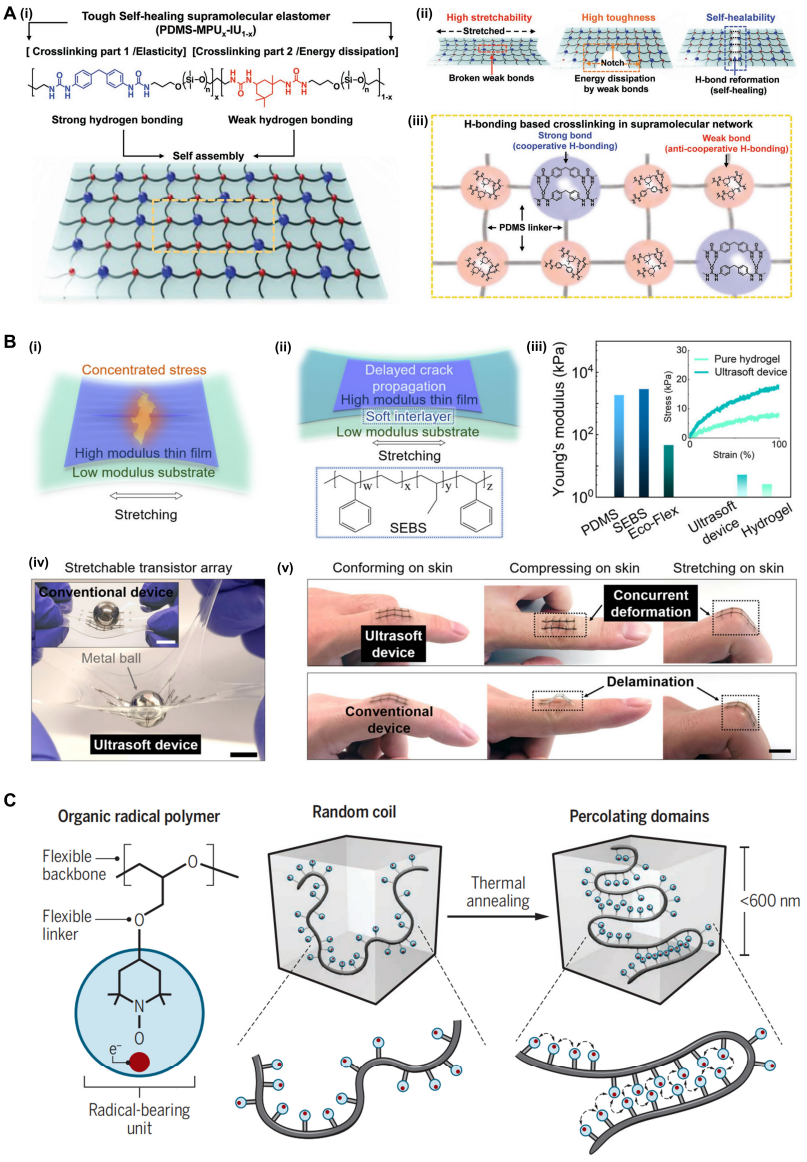


**Figure 2.** (**A**) A Molecular design of the supramolecular elastomer with high toughness, stretchability, and self-healing properties. (i) Chemical structure of PDMS–MPU$_x$–IU$_{1-x}$ and the proposed ideal

supramolecular structure. (ii) Schematics of a stretched polymer film (left), notched film (middle), and healed film (right). (iii) Possible hydrogen bonding combinations for strong bonds and weak bonds, respectively. Adapted from Ref. [84] with permission. Copyright 2018 Wiley–VCH GmbH. (**B**) (i) Sacrificed stretchability when electronic films are stretched on substrates with orders of magnitude lower modulus. (ii) Soft interlayer design uses films with intermediate modulus to reduce the modulus difference at the interface and enable the functional layer to achieve high stretchability. (iii) Young's modulus of ultra-soft and stretchable transistor arrays built on PAAm hydrogel substrates. (iv) Visualization of softer properties of stretchable transistor arrays built on PAAm hydrogel. (v) Highly consistent and imperceptible connections of the ultra-soft transistor arrays on the knuckles at different bending positions (upward). Scale bar, 10 mm. Adapted from Ref. [85] with permission. Copyright 2023 Springer Nature. (**C**) The flexible structure of the organic radical polymer PTEO and the conductivity of the polymer. Adapted from Ref. [88] with permission. Copyright 2018 American Association for the Advancement of Science.

## 4. Engineering Material Structure

The correlation between the properties of e-polymers and the structure of engineered materials is intricate and encompasses a variety of factors. The properties of e-polymers are closely tied to the material's chemical composition, chain organization, morphology, film-processing techniques, interfaces with electrodes, and molecular weight [91]. Understanding and controlling these factors allows for the design of e-polymers with tailored electronic properties. Property–Structure Relationships: The structure of a material directly influences its properties, such as mechanical, electrical, thermal, and chemical characteristics. By studying this structure, researchers can gain a deeper understanding of the basic mechanisms that control these characteristics. Therefore, studying the structure of engineering materials is crucial for understanding the performance structure relationship, optimizing material processing, improving performance and reliability, guiding material selection and design, investigating faults, and promoting material innovation.

Although wearable and implantable bioelectronics have achieved remarkable success and a huge market, their development so far has almost entirely depended on silicon microelectronics, and they have some inherent limitations in providing functions with long-term stability and sustainability [92–94]. The repeated stimulation and damage of these rigid devices to biological tissues often lead to significant inflammatory reactions at the implant site, ultimately leading to human rejection of the device. In addition, the limited lifespan of the implanted power supply in the human body further limits it. Essentially, the difficulties of the interface between these electrons and biological systems stem from the complexity and subtlety of biological systems composed of soft, dynamic, 3D [95–97], and fragile tissues. In addition, animals/humans have innate immunity and are immune to external "invaders." Therefore, we believe that the research of 3D engineering material structure plays a crucial role in achieving the ideal wearable and implantable bioelectronics of stable and sustainable operation in the human body [98–100].

Although the ability to bend can be effectively integrated into small areas or simple curved areas of the body, the complex texture and natural and complex movements of the skin cannot be adapted solely through bending [101–105]. Therefore, the study of stretchability, 3D structures, and other types of structures is crucial. The composition and structural design of the device play a crucial role in the seamless integration of wearable or implantable devices into the human body [106,107]. Mechanical properties play a vital role in determining the quality and durability of a product. By understanding and optimizing these properties, it is ensured that products achieve the highest standards of excellence and provide the highest possible performance [108–110]. Typically, micron/nanofabrication manufacturing processes allow for the large-scale collection of thin films, silicon wafers, and conductive nanofilms/strips/wires [111]. Through this machining process, the corresponding mechanical force exerted on the planar structure of the material transforms the structural shape of the planar material into a 3D structure as a result of the nonlinear buckling process.

### *4.1. 3D Structure*

The human body is a complex 3D structure composed of numerous interconnected systems that maintain homeostasis and perform various functions. Understanding the 3D structure of the human body is crucial for participating in the development of new treatment methods, medical equipment, and technologies to improve health. One way to visualize the 3D structure of the human body is through medical imaging techniques such as computed tomographies (CTs), magnetic resonance imaging (MRI), and ultrasounds. These techniques generate detailed images of internal structures such as bones, organs, and tissues, allowing medical professionals to diagnose conditions, plan surgeries, and monitor treatments. Another way to study the 3D structure of the human body is through anatomical models and simulations. Anatomical models can be physical objects or computer-generated graphics that accurately represent the size, shape, and location of internal structures.

A recently developed compression-buckling method can convert 2D patterns into 3D structures with different geometric shapes. Devices utilizing this 3D curved structure include those based on resistance, capacitance, and piezoelectric effects. In a study, Ryu et al. introduced the method of converting 2D patterns of thin film materials into 3D structures, which has created many exciting opportunities in microsystem design (Figure 3A) [112]. There is a growing area of interest in multifunctional electrical, thermal, chemical, and optical interfaces towards biological tissues, especially 3D multicellular, and milli/micrometer-scale structures, such as spheroids, assemblies, and organoids. The simple and useful functions of compatible and transparent 3D multifunctional mesoscale frameworks (3D MMFs) was introduced in the mechanical characterization of spatially constrained organs. Thin ribbons of parylene-C form the basis of transparent, highly compatible frameworks that can be reversibly opened and closed to capture. The lateral stretching of a thin elastic substrate provides a convenient means for opening and closing these 3D structures. The buckling design in a 3D framework produces a highly spherical cavity whose size can accurately match the corresponding organoid. Finite element technologies can serve as a design tool to guide the selection of geometric shapes and material parameters, as well as the customization of organs required for the shape matching of 3D structures.

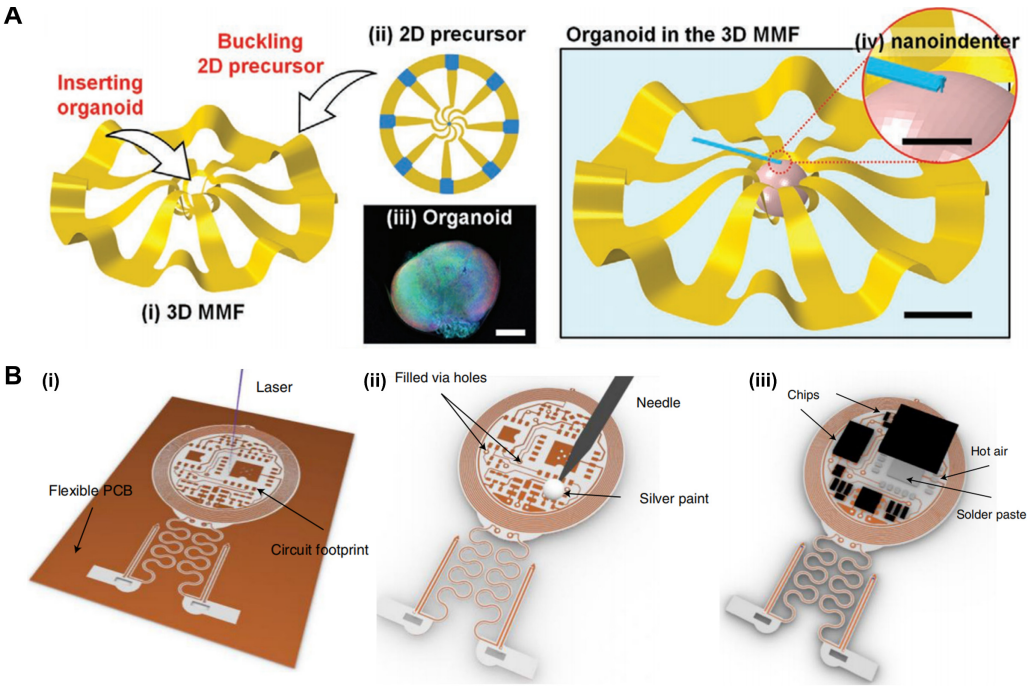


**Figure 3.** *Cont.*

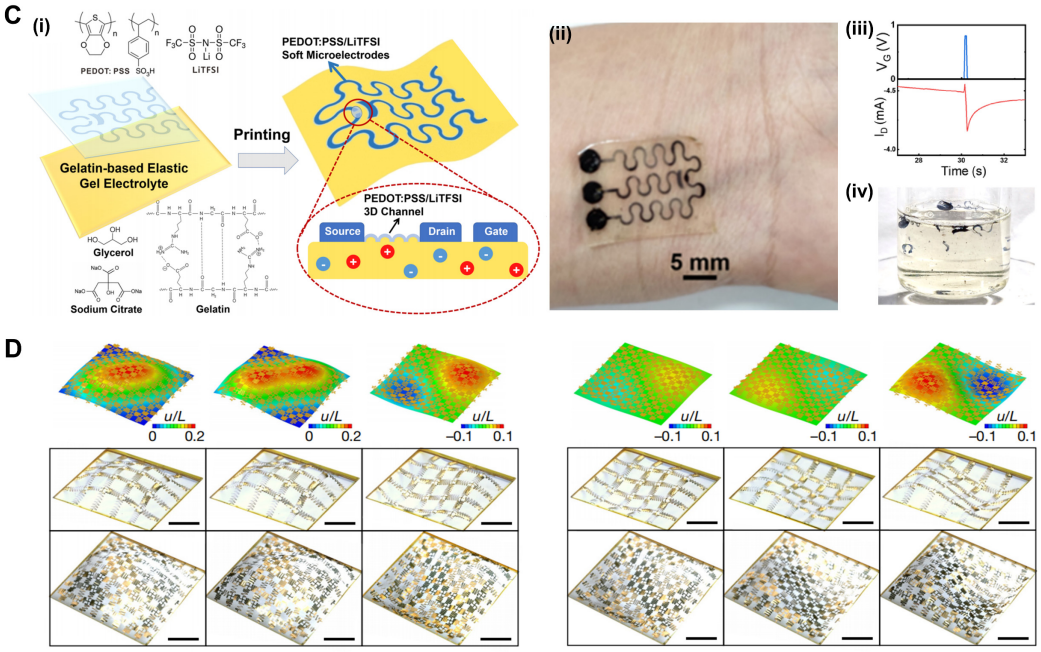


**Figure 3.** (**A**) A Schematic image of an organoid in a 3D MMF. Scale bar: 2 mm. (i) 3D MMF after buckling. (ii) 2D precursor, with bonding sites indicated in blue. (iii) Immunohistochemistry for F-actin (green), NeuN (red), and Nuclear (blue) in a cerebral organoid. Scale bar: 0.5 mm. (iv) Location of nanoindenter on an organoid for indentation. Scale bar: 0.5 mm. Adapted from Ref. [112] with permission. Copyright 2021 Wiley–VCH GmbH. (**B**) HM Device Preparation Flowchart. (i) The polyethylene device is processed by laser-machining technique. (ii) A dull needle is used to dip the laser ablation holes in silver paint. (iii) Connect the electronic components to the circuit substrate. Adapted from Ref. [113] with permission. Copyright 2022 Springer Nature. (**C**) (i) Schematic of a substrate-less all-polymer electromechanical chemical transistor with an active channel layer with a 3D microengineering interface. (ii) Images show that the printed all-polymer OECT can be body-attachable. (iii) Synapse-mimicking with slower current recovery after a voltage spike. (iv) Readily recyclable with water. Adapted from Ref. [114] with permission. Copyright 2022 Springer Nature. (**D**) Finite element analysis and experimental results of the process of deforming the sample into four target abstract shape changes. Scale bars, 5 mm. Adapted from Ref. [115] with permission. Copyright 2022 Springer Nature.

### *4.2. Serpentine Structure*

In consideration of realizing highly stretchable electronic circuits, serpentine structures with stretchable and bendable configurations, such as fractal designs, are used. Rigid conductive films with a planar layout are often bonded or embedded with an elastic substrate to accommodate large strains. The extensibility of the serpentine structure results from the extension of its sinuous "spring" structure. Through the design of serpentine patterns, continuous electronic connectivity can be realized due to the combination of large deformation of flexible elastic substrates and serpentine photolithographic patterning (Figure 3B) [113]. The strategy of such a design of the serpentine pattern enables real-time independent control of the optical stimulus via near-field communication.

### *4.3. Microstructure*

A micromode is a microscale structure incorporating sensor-sensitive components to control and enhance response characteristics. In a study, Wang et al. proposed a generalized strategy for the rapid generation of highly elastic all-polymer OECTs with high transconductance, mechanical stability, and sustainability (Figure 3C) [114]. The ability to achieve such high-performance OECTs was achieved by transferring the microstructures followed by printing on elastic gelatin-based gel electrolytes. In the elastic gel electrolyte,

different active channels are used to produce consumption-mode and enhancement-mode OECTs. It is worth noting that the complex 3D structure of PEDOT: PSS has been designed with imprinted 3D microstructure channels/electrolyte interfaces combined with folded electrodes.

### *4.4. Mesh and Fiber Structure*

The mesh and fiber structure creates softness, flexibility, breathability, and durability characteristics. Functional materials in mesh and fiber structures can be designed as textiles for wearable physiological monitoring. The ultra-thin, porous, and open mesh layout also allows sensors to attach to the skin comfortably or imperceptibly. In a study, Bai et al. [115], demonstrated a mechanical metasurface that is constructed from a matrix of multiple metal trajectories driven with reprogrammable distributed Lorentzian forces that generate a static magnetic field as current passes through (Figure 3D). They passed through the current existing in the static magnetic field. A material structure with an optimized grid of planar conductive properties and programmable control of current distribution provides exciting opportunities. The obtained system exhibits complex and dynamic deformation capabilities, with a response time of within 0.1 s.

## 5. E-Polymers for Bio-Integrated Applications

To ensure human health and safety, emerging wearable devices are biocompatible and ensure a non-irritating interface to allow direct contact with the human body [116,117]. Interface connections to other parts of the body ensure that the interface material is highly breathable, non-toxic, lightweight, and has an elastic and low modulus mechanical response. The human epidermis is a noteworthy interface point for physiological monitoring, and skin bioelectronics is considered an ideal platform for personalized healthcare. Skin bioelectronic devices for long-term, continuous health monitoring provide a robust analysis of various health states, offering access to early disease diagnosis and treatment [118,119]. Traditional rigid silicon microelectronic-based implantable devices have low biocompatibility and high invasiveness [120,121]. In addition, the need for a more sustainable power supply and wireless data transmission options further limits the sustainable development of devices. In the past decade, significant research progress has been made in creating new material concepts and equipment engineering strategies to achieve multifaceted physical and chemical biocompatibility, sustainable power supply, and wireless data transmission under implantation [116,118,122]. Recent chemical and biological strategies have enabled traditional rigid polymer semiconductors to be stretched without affecting their electrical properties. Stretchable pressure sensors are essential for sensing the physical interactions that occur on flexible or deformable skin present in a human body, prosthetic limb, or soft robot. Such sensors have tissue-matched physical and chemical properties as well as wireless communication capabilities with external systems [93,123].

Flexible wearable and implantable sensors are innovative devices that have gained significant attention in the fields of healthcare and fitness, as well as various other industries [116]. These sensors are designed to be worn or implanted on or within the human body to monitor various physiological parameters, environmental factors, and more [124]. They offer several advantages, such as continuous and non-invasive data collection, real-time monitoring, and enhanced comfort for users [125–128]. This section discusses biosensors, pressure sensors, and recently developed devices for the continuous and real-time monitoring of crucial physiological health parameters [117]. The combination of electronics and biological systems has produced many powerful technologies for developing biomedical science [129,130]. The most advanced electrophysiological skin-integrated sensor combines the ultra-thin conformal electrode interface with wireless communication capabilities and low-power electronic devices, suitable for long-term monitoring. Irritation-free direct contact with the skin is ensured by optimally selecting the shape mechanism and material composition components of the skin-like flexible electrodes [131,132].

The integration of energy harvesters with sensors is particularly useful in applications that require long-term or remote monitoring. In remote environmental monitoring stations, these systems can continuously power sensors without the need for frequent battery changes. Additionally, in wearable devices and IoT applications, energy harvesters can extend life and reduce maintenance. Energy harvesters and sensors are two key components in the development of self-sustaining and autonomous systems. Energy harvesters are devices that capture and convert energy from various sources in the environment, while sensors are devices that monitor and measure physical or environmental parameters. These technologies combine to create self-powered sensing systems with a wide range of applications. Self-powered smart sensors are devices that both sense and process data and are self-sufficient in power. These sensors can harvest energy from the environment to sustain their operation, eliminating the need for constant external power sources such as batteries or wired connections.

### *5.1. Pressure Sensor*

Biological interface pressure sensors have different working mechanisms and representative materials used for these sensors [116,121]. For each mechanism, selecting the appropriate material requires balancing sensitivity, stability, flexibility, manufacturability, and many other factors [133–135]. According to different working mechanisms, there are resistive, capacitive, piezoelectric, and triboelectric pressure sensors [136]. E-polymers have gained considerable attention in recent years due to their wide range of applications in the field of pressure sensing.

Scalable pressure sensors are necessary for sensing physical interactions occurring on soft/deformable skin in human bodies [137,138], prosthetics, or soft robots [119]. However, existing types of scalable pressure sensors have inherent limitations, namely the interference of stretching on pressure-sensing accuracy. In a study, Su et al. proposed a highly scalable and sensitive pressure sensor design that can provide unchanged sensing performance under tensile conditions (Figure 4A) [139]. This is achieved through the collaborative creation of an ion-capacitive-sensing mechanism and layered mechanical microstructure. Through this optimized structure, this sensor exhibits 98% strain insensitivity up to 50% strain and 0.2 Pa. This sensor has been used to achieve precise sensations of physical interaction on the skin of humans or soft robots. In a study, Chen et al. reported cellulose ion-conductive hydrogel (ICH) rationally designed from both nano and micron perspectives for ultrasensitive pressure sensors (Figure 4B) [140]. By introducing low-molecular weight cellulose and utilizing the idea of rough surfaces, the piezoelectric capacitance sensitivity of the brain chip has increased from 0.04 $kPa^{-1}$ to 89.81 $kPa^{-1}$, exhibiting high transparency, excellent durability, and good electrical transmission. In addition, brain chips are used in sensors and arrays, including medical and motion recognition. This design is also applicable to piezoresistive tactile sensors, achieving improved sensitivity. This cost-effective, effective, and environmentally friendly technology undoubtedly provides a new perspective and the potential to enhance the functionality of flexible pressure sensors. E-polymers offer several advantages for pressure sensing applications. They are lightweight, flexible, and can conform to irregular shapes, making them suitable for a range of form factors. Additionally, they can be processed using low-cost fabrication techniques, such as solution-based deposition or printing methods, enabling the large-scale production of sensors. E-polymers, including PEDOT, PPy, and PANI, are promising materials for pressure-sensor applications. Their unique combination of electrical and mechanical properties makes them suitable for developing flexible, lightweight, and sensitive sensors for various applications, including wearable devices, robotics, and biomedical sensing.

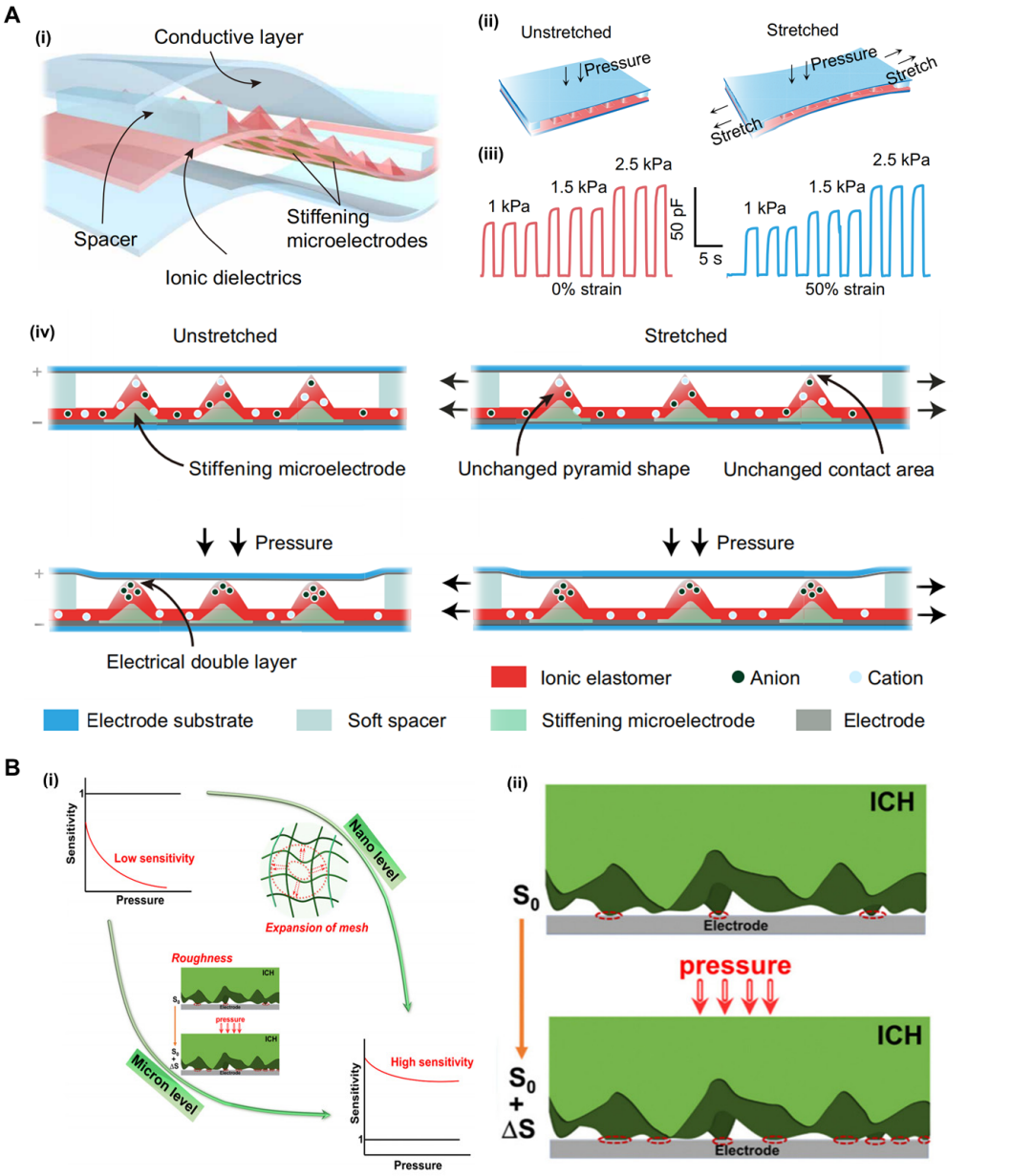


**Figure 4.** (**A**) A stretchable and strain-unperturbed pressure sensor for motion interference-free tactile monitoring on skins. (i) The 3D structure of sensors. (ii) Schematic diagram of the pressure sensor in the unstretched (left) and stretched (right) states. (iii) Capacitive response for three repetitive pressure values. (iv) Schematic diagram of a pressure-sensing mechanism that maintains constant performance under tensile conditions. Adapted from Ref. [139] with permission. Copyright 2021 American Association for the Advancement of Science. (**B**) Rationally designed cellulose hydrogel for an ultrasensitive pressure sensor. (i) Design and regulation of cellulose hydrogel for boosting sensitivity at the micron and nano levels. (ii) Mechanistic diagram of a rough sample under pressure. Adapted from Ref. [140] with permission. Copyright 2023 Royal Society of Chemistry.

### *5.2. Biosensor*

E-polymers are attracting attention in the field of biosensors due to their unique properties that can be used in sensing applications. A biosensor is an analytical device that combines a biosensing element with a sensor to detect and analyze the presence of a specific biological target [141,142]. E-polymers can be used as transducer elements in biosensor platforms to convert the interaction between the biosensing element and the target analyte into a measurable electrical signal. Conducting polymers such as polyaniline

(PANI), PPy, polythiophene (PTH), and poly(3,4-ethylenedioxythiophene) (PEDOT), which are highly conductive, have been extensively studied for biosensor applications [143–145]. In addition, molecularly imprinted polymers (MIPs) selectively recognize and bind to specific target molecules, and MIPs can be used as recognition elements in biosensors for the detection of a wide range of analytes, including small molecules, proteins, and even whole cells [146]. Polymer hydrogels are highly biocompatible and capable of encapsulating and immobilizing biomolecules. Through the addition of specific receptors or enzymes to the hydrogel matrix, the swelling or shrinking of the hydrogel can be converted into an electrical signal, indicating the presence of an analyte [147]. This section focuses on the use of OFETs, which utilize an organic semiconductor polymer as the active ingredient in the transistor structure and can provide quantitative information about the target analyte by measuring changes in the electrical properties of the organic semiconductor.

The choice of polymer depends on the specific application requirements, including target analytes, sensitivity, selectivity, and compatibility with biological systems. OECTs with uniquely high amplification and biosignal sensitivity are novel device platforms for next-generation bioelectronics [121]. OECTs are particularly interesting due to their biocompatibility and ability to efficiently interface with biological systems, making them useful in applications such as bioelectronics and medical devices. OECTs are made using organic materials, such as organic semiconductors and organic electrolytes. These organic materials offer several advantages, including flexibility, biocompatibility, and ease of manufacturing [148]. OECTs have unique properties that make them well-suited for applications where biocompatibility, flexibility, and precise control of electrical conductivity are essential. They continue to be an active area of research and development in the field of organic electronics and bioelectronics [53,149]. In a study, Dai et al. discovered a stretchable semiconductor (p(g2T–T)) for OECT devices with high strain, repeatable stretching, and OECT performance comparable to state-of-the-art devices (Figure 5A) [150]. The key design features of this polymer are a nonlinear main chain structure, moderate side chain density, and sufficiently high molecular weight. This polymer semiconductor with high stretchability prepared an OECT with both high electrical conductivity and biaxial stretchability. In addition, the electrocardiogram (ECG) records on the skin were also displayed, combined with the height of the skin. In a study, Li et al. synthesized an e-polymer that is a bioadhesive polymer semiconductor, a dual network structure formed by a bioadhesive polymer, and a redox-active semiconductor polymer [151]. The semiconductor thickness films prepared from this e-polymer can be quickly and firmly adhered to the tissue surface and exhibit excellent properties, including good biocompatibility, high carrier mobility, and high stretchability. As a result of the excellent properties of this material, a fully bioadhesive transistorized sensor was further fabricated. The results show that this sensor is capable of stable electrophysiological recording of isolated rat hearts and living rat muscles, and the demonstrated recording values are of high quality (Figure 5B). In a study, Qing et al. discovered that the OECT biosensor could be adaptively, sustainably, and stably implemented via thermoelectric fabric (TEF) without any additional accessories (Figure 5C) [152]. In practical applications, an all-fiber integrated thermoelectrically powered physiological monitoring device (FPMD) in vivo generates a persistent and stable output signal and displays a linear monitoring region of glucose in artificial sweat (sensitivity 30.4 NCR (normalized current response)/dec, 10–50 μM), which has reliable performance in anti-interference and reproducibility. This device can be extended to monitor various biomarkers and provides a new strategy for building wearable, comfortable, highly integrated, and self-powered biosensors.

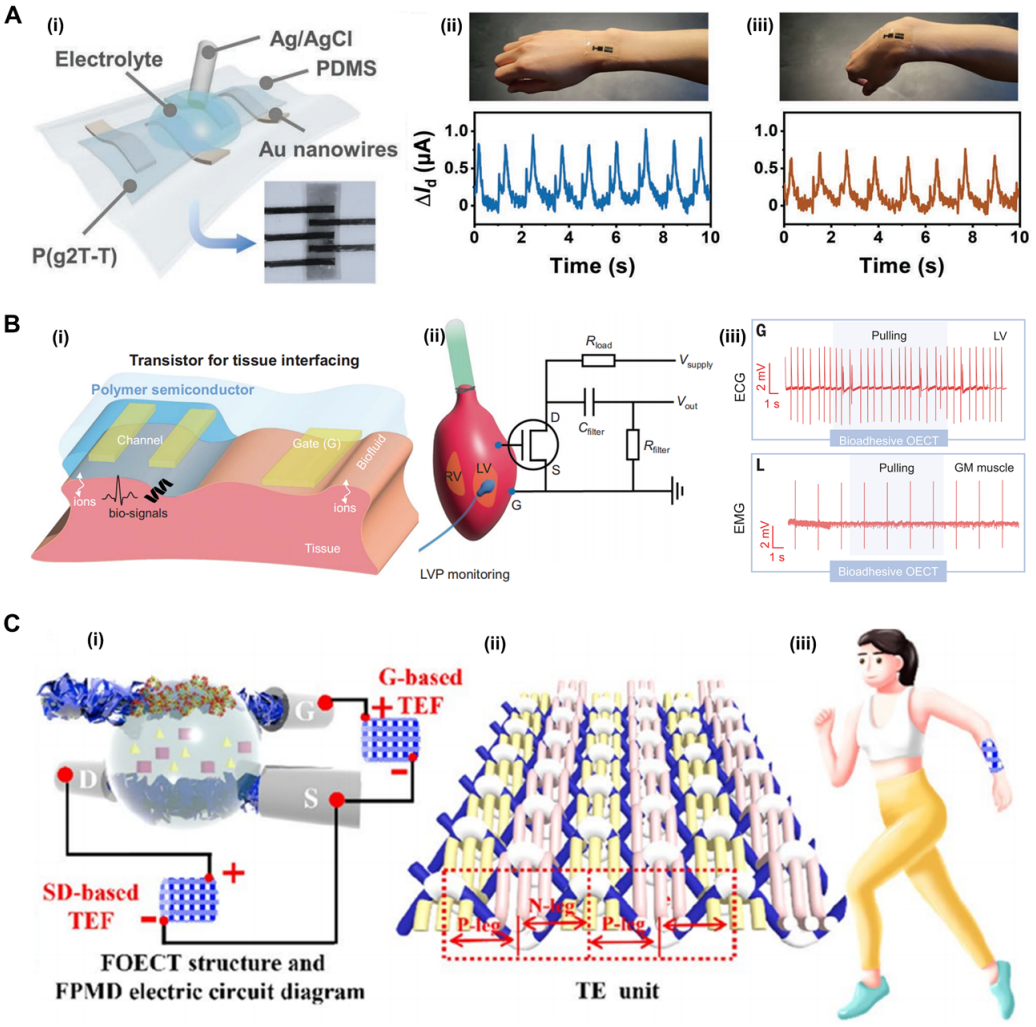


**Figure 5.** (**A**) P(g2T–T)-based intrinsically stretchable OECT and its function as a wearable ECG sensor. (i) Schematic showing the structure of the intrinsically stretchable OECT. (ii, iii) Photographs showing the conformable attachment of the OECT on the wrist without (ii) and with (iii) bending, and the corresponding ECG signals acquired from the human body. Adapted from Ref. [150] with permission. Copyright 2022 Wiley–VCH GmbH. (**B**) Stretchable OECT based on P(g2T–T) material and as a wearable ECG sensor. (i) Schematic diagram of biosensing using electrochemical transistors at the tissue interface of a living organism. (ii) Schematic and circuit diagram of a sensor recording ECG on the surface of the heart. (iii) ECG signals recorded by left ventricular total bio-adhesion OECT and EMG signals recorded by GM muscle total bio-adhesion OECT. Adapted from Ref. [151] with permission. Copyright 2023 American Association for the Advancement of Science. (**C**) (i) Enlarged view of fiber-assembled OECT based on Cotton/PDG and FPMD circuit diagram where terminals between TEF and fiber-assembled OECT are connected by flexible wires. (ii) Enlarged view of the TE unit in the TEF. (iii) Diagram of sensing application of FPMD. Adapted from Ref. [152] with permission. Copyright 2023 Springer Nature.

### *5.3. Energy Harvester*

Traditional battery power supply has disadvantages such as limited energy supply life, complex packaging process, easy leakage, severe toxicity and pollution, fixed size, and high cost [153,154]. Therefore, using energy collectors instead of batteries achieves a self-powered system [155]. Mechanical vibration energy is a common form of energy in the environment, which exhibits a more sustained, stable, and high-density energy than solar and thermal energy [156–158]. Future electronic products will have flexibility and deformability while maintaining the flexibility and deformability of their power supply, which

is also very important [159,160]. The stretchable deformation triboelectric/piezoelectric nanogenerator can collect biomechanical energy as the power supply of wearable electronic products. Triboelectric generators operate mainly in contact mode and sliding mode, and their efficiency depends strongly on the differences in the constituent materials' electron attraction capacity and the contact surfaces' morphology [161,162]. Triboelectric devices can utilize a wider range of materials (PTFE, PET, PI, PDMS, PMMA, CNT, ITO, Al, Cu, Si, etc.). Higher output power densities and energy-conversion efficiencies can be realized depending on the choice of material and the design of the structure, for example [152]. A piezoelectric nanogenerator is a device that uses materials with a piezoelectric effect to convert mechanical energy into electrical energy to supply power for nanodevices when subjected to external tension or compression [162–164].

E-polymers have gained significant attention in the field of energy harvesting due to their unique properties and potential applications. In a study, Zhang et al. utilized a lignosulfonate (LS)–$Al^{3+}$ composite system to rapidly prepare a PAA ion hydrogel and used it to obtain electrical energy from atmospheric water (Figure 6A) [165]. The semi-quinone radicals generated by the oxidation of LS by ammonium persulfate (APS) can quickly initiate the polymerization of acrylic monomers into PAA polymer chains. Then, the green crosslinker $Al^{3+}$ ion can crosslink the polymer chain into a hydrogel through ion interaction within a few minutes at room temperature. The excellent mechanical and adhesive properties make it very easy to assemble hydrogels into a low-cost, flexible, and efficient hydrogel-based moisture generator (HMEG). HMEG also exhibits good output stability, long-term output durability, and all-weather workability under mechanical deformation conditions. In a study, Bian et al. developed an all-organic triboelectric nanogenerator (TENG) based on a 3D PPy network, which enabled energy harvesting and bionic pressure sensing (Figure 6B) [166]. By combining with polydimethylsiloxane (PDMS) and barium titanate nanoparticles, this composite triboelectric interface not only has high uniaxial tensile strength but also has a high short-circuit current density that responds to environmental or human motion. This excellent electrical response further enables it to detect environmental mechanical energy sensitively. These results demonstrate an ideal method for developing self-driving electronic skin and biomimetic tactile sensors, which have interesting elasticity, stretchability, and sustainability. Electrode-conductive materials are key components of nanogenerators due to their low cost, large production scale, simple synthesis process, and controllable conductivity. Implantable enzyme-enabled biofuel cells (BFCs), as an effective and sustainable alternative to energy storage devices, can utilize enzymes (e.g., glucose oxidase ($GO_X$) and lactate oxidase ($LO_X$)) as catalysts to convert biofuels such as glucose, urea, and lactate into bioelectricity. In a study, Guan et al. demonstrated a stretchable and flexible electrostatic spinning-based glucose/oxygen biofuel cell with a glucose oxidase bioanode, a Pt/C cathode, and a thermoplastic polyurethane substrate capable of adapting to a variety of stresses and strains induced by individual motions (Figure 6C) [167]. Stable covalent connections between activated carbon nanotubes and glucose oxidase within a three-dimensional thermoplastic polyurethane network were established via an amide reaction to ensure rapid direct electron transfer at the bioanode and stable power output of the device in a flexible environment. Therefore, e-polymers have been widely used in energy harvesting science. E-polymers are also becoming increasingly important in the field of energy harvesting. The field of e-polymers for energy harvesting is still growing rapidly. With discoveries and advancements, e-polymers will likely play an important role in the development of future energy-harvesting technologies.

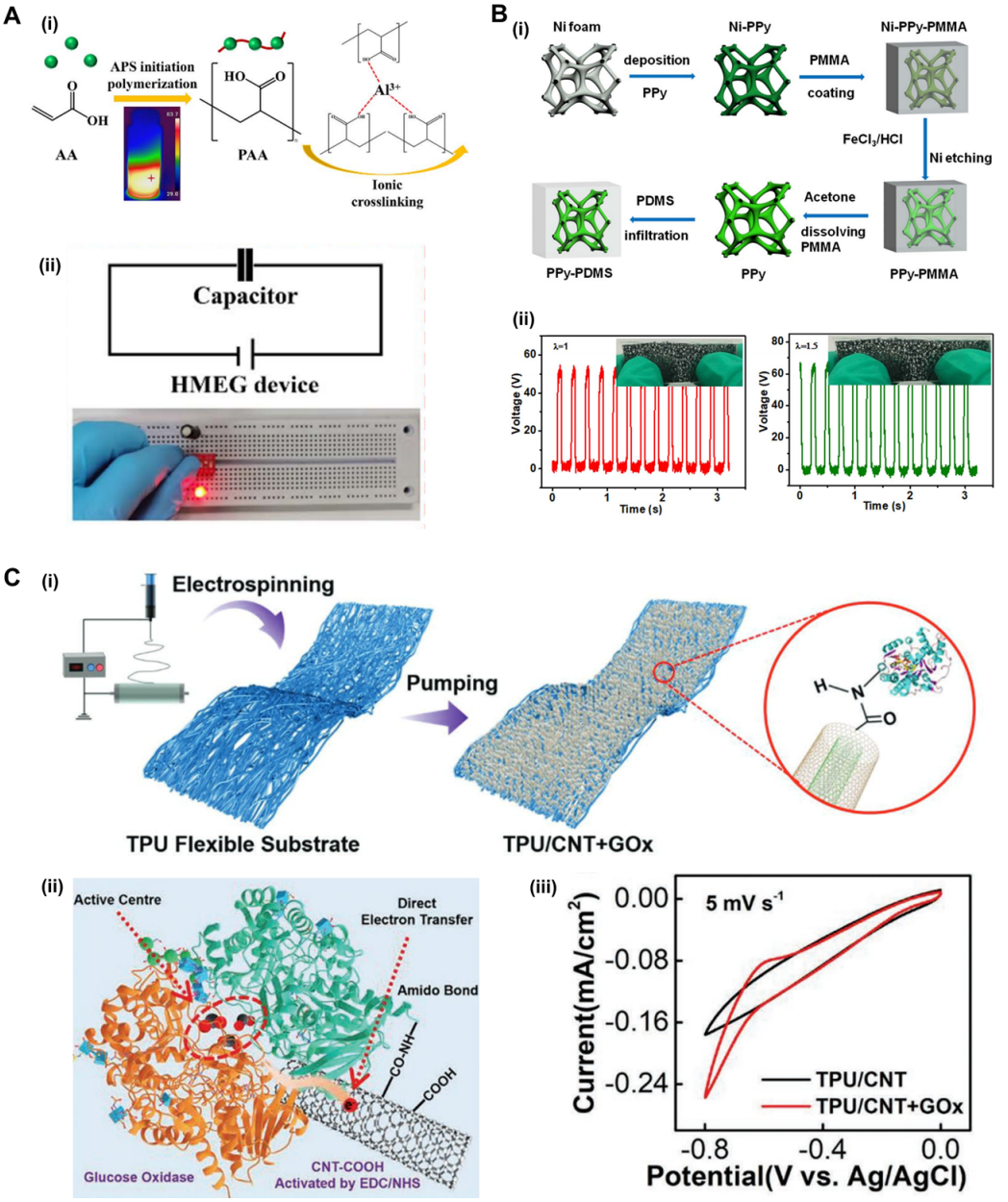


**Figure 6.** (**A**) (i) Schematic of the LS–$Al^{3+}$-PAA hydrogel synthesis. (ii) Photograph of a red LED bulb powered by the capacitors charged by the series-connected HMEGs. Adapted from Ref. [165] with permission. Copyright 2023 Royal Society of Chemistry. (**B**) (i) Schematic synthesis of the PPy–PDMS foam. (ii), Corresponding output voltage and current of the P–TENG. Adapted from Ref. [166] with permission. Copyright 2018 Elsevier Ltd. (**C**) (i) Schematic illustration of the detailed synthetic procedure for TPU/CNT+$GO_X$. The circle highlights the immobilization of $GO_X$ via the functional groups. (ii) Schematic illustration of direct electron transfer between $GO_X$ active center and CNT. (iii) CV curves of TPU/CNT+$GO_X$ and TPU/CNT in 0.1 M PBS. Adapted from Ref. [167] with permission. Copyright 2023 Wiley–VCH GmbH.

### *5.4. Strategies for Self-Powered Intelligent Sensing Systems*

Combining sustainable electricity and reliable signal sensing has brought challenges and opportunities to electronic systems in the Internet of Things era [95]. In the new generation of the Internet of Things, collecting and analyzing big data based on widely distributed perception networks is particularly important in developing intelligent systems. Conventional sensors usually require an external power supply but have a limited lifetime and high maintenance costs. In a study, Ohayon et al. demonstrated an integration of n-type

conjugated polymers with oxidoreductases to automatically detect glucose and humoral electricity generation (Figure 7) [168]. A reversible, dielectric-free miniature glucose sensor is an enzyme-coupled electrochemical transistor with a detection range of up to six orders of magnitude. This n-type polymer can also be used as the anode of an enzyme fuel cell and paired with a polymer cathode to convert chemical energy from glucose and oxygen into electrical energy. All polymer biofuel cells were glucose in solution and stable for more than 30 days. As a newly developed mechanical energy harvesting and mechanical force-sensing technology, TENGs have great potential to overcome these limitations [169]. Most importantly, TENGs can be manufactured from wood, paper, fibers, and polymers, the most commonly used materials [170]. In a study, Lai et al. introduced a responsive soft robot that is enabled by a highly scalable and sensitive self-actuated triboelectric skin, which harvests energy through the triboelectric effect and actively senses proximity, contact, and pressure [171]. Even if the skin of the triboelectric body is stretched to a 100% strain, it can still maintain a sensing function. These compatible features enable tribal skins to integrate into soft robots, actively sensing external stimuli and internal movements through self-generated electrical signals. Self-powered devices can be used not only for sensing, detection and monitoring, but also for driving functional components that interact with humans in a real-time. In addition, soft robots with large-area multi-channel sensing arrays have been proven. This work opens a crucial door to the enormous potential of soft robots and artificial electronic sensory skins.

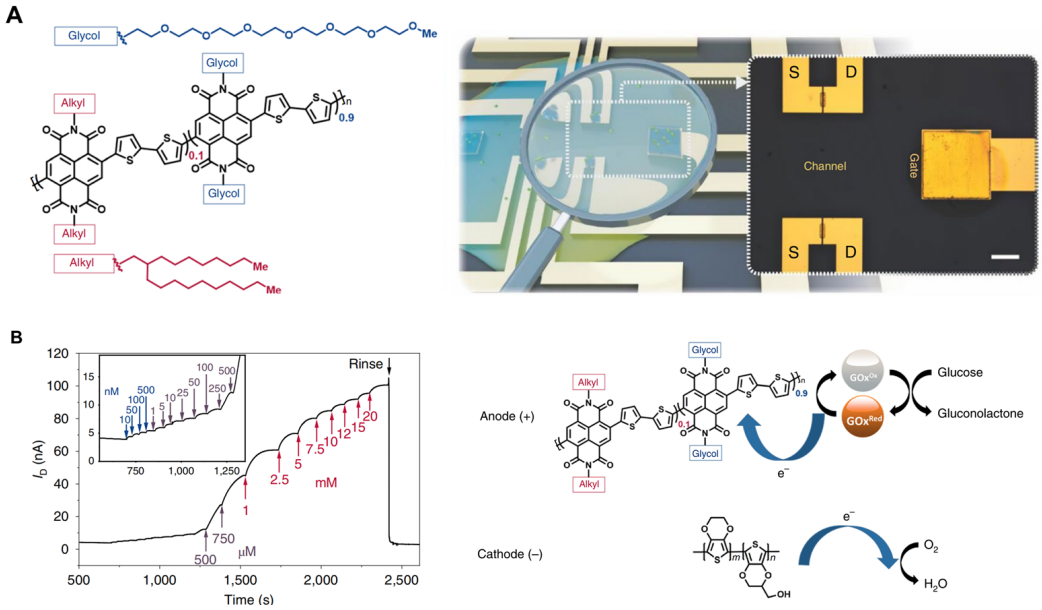


**Figure 7.** (**A**) The chemical structure of the n-type copolymer P-90 (left), schematic illustration of the sensor (right). (**B**) Real-time response of the OECT as successive amounts of glucose were added to the buffer (left), reactions that occur during the operation of the Enzymatic fuel cell (right). Adapted from Ref. [168] with permission. Copyright 2020 Springer Nature.

## 6. Conclusions and Outlook

The progress in the synthesis, properties, engineering material structures, sensing applications, power supply, and system-integration strategies based on e-polymer materials reviewed in this article provides a strong foundation for new skin-like multifunctional wearable systems and implantable human body systems, with broad potential applications in clinical, consumer, and research fields. E-polymers have greatly increased their functionality in clinical medicine, aesthetics, and digital health beyond the capabilities of traditional electronic systems. Impressive progress has been made in translating research results into practical applications, but many significant and interesting challenges still exist.

Research on wearable and implantable sensors is a technology with strong potential for personalized healthcare and continuous monitoring of human physiological health conditions. The most recently developed biosensors are also currently based only on material–chemical detection schemes that fulfill the requirements of physical interface systems. As a result, the existing libraries of active electronics, biochemical conductivity, encapsulation layers, and flexible stretchable materials continue to expand. One of the most influential efforts in these areas is focused on supporting the requirements of biotic/non-biotic functional interfaces. In addition, different populations may have certain differences due to physiological issues that result in fundamental incompatibility with wearable systems. Consequently, the requirement to develop sensor devices for biochemical and environmental signals is also essential.

The limitations of power supplies have become a critical concern that demands renewed attention today, particularly to the size, weight, and form factor of power supplies. Advances in energy harvesting technology are paramount, for which continued progress and perhaps new directions are desirable to support the growing demands regarding operating distance, sampling frequency, communication bandwidth, and lifetime. Energy harvesting offers the potential for self-powered operation of smart sensors. TENGs can be integrated with specific passive sensors to enable self-powered, multifunctional sensing systems by harvesting energy from the mechanical motion as a power source. TENG-based self-powered charging systems can also harvest energy from movements as a sustainable power source.

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