

E-Polymers

Subjects: Polymer Science

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E-polymers, also known as conducting polymers, are a class of materials that exhibit both electrical conductivity and the mechanical properties of polymers. The use of e-polymer materials in daily life is becoming increasingly widespread, especially in the field of biology. 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. 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, thereby reducing the possibility of any long-term complications. 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.

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

1. 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 ^[1]. 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 ^[2]. E-polymers utilize specialized chemical substances and composite materials for biological applications ^{[3][4]}. 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 ^{[5][6]}. 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 ^{[7][8]}. 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 ^{[9][10]}. 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 ^{[9][11][12]}. 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 ^[13]. 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 ^[14]. 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 ^[15]. 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 [16]. 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 [17][18]. For the construction of CMOS logic circuits and p–n junction devices, n-type semiconductors play a crucial role [19]. Among various electronic deficient components, cyanide-functionalized hydrocarbons are emerging to achieve high-performance n-type organic and polymer semiconductors [20]. 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 [21]. 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 [22]. Consequently, transistor arrays essentially constitute stretchable skin electronics, including active matrices for sensing arrays as well as analog and digital circuit elements.

2. 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 [22][23][24]. 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 [25][26]. 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 [27][28][29]. 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 [30][31][32]. 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 [33]. 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 [34]. 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 [35]. 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 [36].

A growing and widespread concern is the application of e-polymers to bio-interfaces and organisms [37][38]. 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 [39]. 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 [40]. Due to 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 [41][42]. The electrical conductivity of organic radical polymers is much higher than expected, and organic radical polymers have unusual electronic properties [43]. 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 [44][45]. Therefore, it will be important to find suitable doping agents and dope them reasonably with conductive polymers.

3. 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 [46]. 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 [47]. 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 electronics and biological systems stem from the complexity and subtlety of biological systems composed of soft, dynamic, 3D [48][49], 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 [50]. 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 [51]. 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 [52]. 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 [53]. Typically, micron/nanofabrication manufacturing processes allow for the large-scale collection of thin films, silicon wafers, and conductive nanofilms/strips/wires [54]. 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.

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. 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 strategy of such a design of the serpentine pattern enables real-time independent control of the optical stimulus via near-field communication. A micromode is a microscale structure incorporating sensor-sensitive components to control and enhance response characteristics. 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 [55].

4. 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 [56]. 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 [57][58]. Traditional rigid silicon microelectronic-based implantable devices have low biocompatibility and high invasiveness [59]. 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 [60]. 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 [61].

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. These sensors are designed to be worn or implanted on or within the human body to monitor various physiological parameters, environmental factors, and more [62]. They offer several advantages, such as continuous and non-invasive data collection, real-time monitoring, and enhanced comfort for users [63]. This section discusses biosensors, pressure sensors, and recently developed devices for the continuous and real-time monitoring of crucial physiological health parameters. The combination of electronics and biological systems has produced many powerful technologies for developing biomedical science [64]. 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 [65].

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.

4.1. Pressure Sensor

Biological interface pressure sensors have different working mechanisms and representative materials used for these sensors [66]. For each mechanism, selecting the appropriate material requires balancing sensitivity, stability, flexibility, manufacturability, and many other factors [67]. According to different working mechanisms, there are resistive, capacitive, piezoelectric, and triboelectric pressure sensors [68]. 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 [69], prosthetics, or soft robots. However, existing types of scalable pressure sensors have inherent limitations, namely the interference of stretching on pressure-sensing accuracy. 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.

4.2. 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 [70]. 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 [71]. 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 [72]. 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 [73]. 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. 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 [74]. 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 [75].

4.3. 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 [76]. Therefore, using energy collectors instead of batteries achieves a self-powered system. 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 [77]. Future electronic products will have flexibility and deformability while maintaining the flexibility and deformability of their power supply, which is also very important [78]. 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 [79]. 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. 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 [80]. E-polymers have gained significant attention in the field of energy harvesting due to their unique properties and potential applications. 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.

4.4. 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. 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. As a newly developed mechanical energy harvesting and mechanical force-sensing technology, TENGs have great potential to overcome these limitations. Most importantly, TENGs can be manufactured from wood, paper, fibers, and polymers, the most commonly used materials [81]. 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.

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