MSMEE 2024

Organic Electronics in Flexible Devices: Engineering Strategy, Synthesis, And Application

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Abstract. In recent years, flexible electronics have made significant strides, especially in organic light-emitting diodes, photovoltaics, thin-film transistors, integrated circuits, sensors, and memories. This study investigates the pivotal role of organic electronics in shaping flexible and wearable technology. It explores key aspects such as conductive polymers, small molecule organic semiconductors, and the use of organic dyes and pigments in displays, alongside synthesis techniques like polymerization and vapor deposition. These methods highlight advances in flexibility engineering and integration into wearable devices, enabling innovations like textile-integrated electronics and ultra-thin, skin-like interfaces. The discussion also covers synthesis and fabrication strategies, emphasizing cost-effective, solution-processing techniques for manufacturing electrodes, interconnects, and metal contacts on various flexible substrates. Advanced materials like graphene polymer composites, carbon-nanotube composites, and polymer-ceramic composites are investigated for enhancing stretchability and flexibility in device architectures. Furthermore, the paper examines the evolution of flexible devices, focusing on design strategies for flexibility and stretchability, innovative engineering polymers, composites, and device architectures, including 3D integration. Integration techniques in wearable devices, such as textile integration and skin-like electronics, are discussed, showcasing their transformative potential in applications like health monitoring devices, smart clothing, and wearable displays. Finally, the paper proposed solutions to address challenges such as durability, stability, scalability, and manufacturing efficiency, emphasizing the ongoing quest for improved materials and manufacturing processes. The conclusion underscores the transformative potential of organic electronics, envisioning a new era of wearable and flexible technology seamlessly integrated into daily life, revolutionizing our interactions with electronic devices.

Keywords: Flexible electronics; organic electronics; wearable devices; flexibility engineering.

1. Introduction

In recent years, significant improvements have been achieved in the field of flexible electronics, including organic light-emitting diodes, organic photovoltaics, organic thin-film transistors, organic flexible integrated circuits, sensors, and memories [1]. This investigation explores the critical role that organic electronics have had in the development of flexible and wearable electronics. This paper navigates through the fundamentals of conductive polymers, small molecule organic semiconductors, and the role of organic dyes and pigments in color displays. Synthesis methods, including polymerization, solution processing, and vapor deposition, are discussed, highlighting technological advancements in flexibility engineering and wearable device integration. These innovations have led to the development of textile-integrated electronics and ultra-thin, skin-like interfaces. Practical applications, such as health monitoring devices, smart clothing, and flexible displays, are explored, reshaping user experiences. The rapid advancement of flexible and wearable electronics underscores the importance of low-cost, solution-processing, and high-throughput techniques for producing metal contacts, interconnects, and electrodes on various flexible substrates [2].

2. Organic Materials for Flexible Electronics

2.1. Fundamentals of Organic Materials

Flexible electronics are mostly composed of organic materials because of their special characteristics and adaptability. For example, organic materials are inherently flexible, allowing them to bend and conform to various shapes without breaking. This property is crucial for the development of flexible electronic devices like wearable electronics, rollable displays, and flexible sensors. Besides, Organic materials are often lightweight, making them suitable for applications where weight is a critical factor, such as wearable devices or lightweight displays. The following key aspects will be covered in this section.

2.1.1 Conductive Polymer Qualities

Conductive polymers are crucial components of flexible electronic devices. The electronic properties of conductive polymers can be tuned by modifying their chemical structures. Because of its tunability, conductivity, charge carrier mobility, and other electrical properties can be optimized to better meet the requirements of flexible electronic applications. This section will provide an in-depth exploration of the properties and types of polymers, such as PEDOT: PSS, polyaniline, and polythiophenes.

Poly(3,4-methylenedioxy-thiophene): poly (styrene sulfonic acid) (PEDOT: PSS) is a polymer that is a mixture of two ionomers. One of the mixtures is polystyrene sulfonate, for which part of the sulfonyl groups carry a negative charge. In addition, the other ionomer poly(3,4-ethylenedioxythiophene) carries a positive charge due to the conjugation base on the polythiophene. Therefore, the macromolecule salt consists of two ionomers giving rise to a high conductivity and thermal stability. High electrical conductivity ensures that electrical signals are efficiently transmitted within the device, and thermal stability ensures that electrical components operate within a specific temperature range without degradation or failure. In addition, a stretchable polymer mix based on poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT: PSS) exhibits low sheet resistance [3].

Polyaniline (PANI) is a well-known conductive polymer that has gotten a lot of attention from nanotechnology researchers for its potential to improve sensors, optoelectronic devices, and photonic devices [4]. In the polymer chain of PANI, the alternating arrangement of the benzene rings and the nitrogen atoms allows the PANI to exist a various oxidation state, such as leucoemeraldine base (completely reduced), pernigraniline base (fully oxidised), and emeraldine base(semi-oxidised) [5]. Therefore, the unique electrical and optical characteristics of each oxidation state make polyaniline adapt to a range of applications. The optical absorption properties of PANI can be adjusted according to their molecular structure. This tunability enables the optimization of light absorption in certain wavelength ranges, which is critical for applications such as photodetectors and solar cells embedded in flexible electronics.

Polythiophene and its derivatives have received substantial interest due to its excellent environmental stability, simplicity of structural modifications, and diversity in optical and electrochemical properties [6]. From the molecular structure of the polythiophene, the conjugated backbone structure of polythiophenes is made up of thiophene rings that alternate. Additionally, this conjugation contributes to electronic conductivity by enabling the delocalization of π-electrons along the polymer chain to improve signal integrity by minimizing signal degradation during the transmission.

2.1.2 Small Molecule Organic Semiconductor

The small molecule organic semiconductor plays a significant role in flexible electronic applications. Because small molecules have the potential to form well-ordered crystalline structures. This high degree of crystallinity enhances charge carrier mobility, allowing for efficient electron and hole transport within the semiconductor. Additionally, small-molecule organic semiconductors generally have higher charge carrier mobility than their polymer counterparts. This characteristic helps to achieve faster response times, higher switching speeds, and better overall device performance in flexible electronics. The properties of two main examples (pentacene and perylenetetracarboxylic dianhydride) will be introduced in this section.

Pentacene is a famous conjugated chemical compound, which is widely utilized in electrical devices, including organic thin-film transistors (OTFTs), organic light-emitting diodes (OLEDs), photodetectors, and intelligent sensors [7]. From the structural perspective, pentacene has a planar, π-conjugated structure made up of five linearly fused benzene rings. This shape allows effective electron delocalization, allowing charge carriers to easily flow down the molecular backbone. Because of its comparatively high mobility, pentacene has been used for electronic technology as a p-type organic semiconductor and as a hole transport material [8].

Perylene-3,4,9,10-tetracarboxylic dianhydride (PTCDA) has been extensively studied for decades due to its unique electrical and optical properties, as well as applications in organic electronics, surface engineering, and 2D material passivation [9]. Moreover, in the solid state, PTCDA can form well-ordered crystalline structures with molecules arranged closely together. The ordered molecule packing is essential for efficient charge transport in organic semiconductor devices.

2.1.3 Organic Dyes and Pigments

Organic dyes and pigments are used to create the different colors emitted by OLED displays. These materials allow for the production of vibrant and high-contrast images, enhancing the visual appeal of flexible displays. Moreover, stretchable and flexible polymeric materials can be combined with organic pigments and dyes. This flexibility increases the potential uses in wearable gadgets, curved screens, and flexible electronics. It is also essential for the creation of bendable and rollable displays.

2.2. Synthesis and Fabrication Strategies

Specialized processes are needed for the synthesis and fabrication of organic materials to achieve the desired characteristics. Conductive polymers are synthesized by using polymerization processes, which include chemical and electrochemical approaches [10]. This process gives fine control over the structure and characteristics of the resulting polymers. Additionally, spin-coating and inkjet printing are examples of solution-processing techniques for device fabrication [11,12]. Indeed, Applications for inkjet and 3D printing are numerous and include the fabrication of a wide range of devices designed to improve and simplify the design, fabrication, and functionality of sensors and analytical platforms [12]. Additionally, small molecule semiconductors can be deposited using vapor deposition techniques [13].

3. Technologies advancements in Flexible Devices

The advancements in flexible devices mark the revolution for electrical circuit design, which is particularly highlighted by the unseen versatile integration into bendable and wearable devices. The following section primarily focuses on the material and techniques in the structural design and application of flexible devices, exploring both organic-based flexibility and innovative integration into wearable devices.

3.1. Designing for Flexibility and Stretchability

3.1.1 Different Techniques and Engineering Polymers

As a group of materials, engineering polymers are characterised by their exceptional mechanical or chemical properties, which render them highly suitable for implementation in flexible devices. Besides their flexibility and tensile strength, they retain essential stability in a wide range of environmental conditions.

Several physical or chemical techniques are frequently employed to further adjust their mechanical strength and thermal tolerance. By modifying the sequence of several monomer structures.

co-polymerization enhances the structure to create a more suitable substrate for subsequent modification. For instance, chemically substituting a portion of the ethylene glycol backbone of Polyethylene Terephthalate (PET) with the less structurally conformable cyclohexanedimethanol (CHDM) compromises the degree of orderly crystallisation. This insertion leads to a decrease in the polymer's melting point [14], hence enhancing its processability in shaping. Various substrates can also be physically blended together to demonstrate a collective property. Different proportions of polyimide (PI) blended with polyetherimide (PEI) can be subjected to pyrolysis to create thermally stable carbon molecular sieve (CMS) membranes [15], with the ability to chemically adsorb and desorb carbon dioxide at varying temperatures. To boost mechanical strength, chemically inert fillers could also be incorporated into the polymer structure. For instance, the addition of silica nanoparticles to Polydimethylsiloxane (PDMS) results in a significant increase in tensile strength, specifically by 129% compared to the control material [16]. Chemical modifications on the surface also provide a more precise alignment for the use of the substrate. By adding the polar hydroxy group (-OH) to the initially hydrophobic PDMS surfaces, the amount of hydrophobic protein sticking to the surface is decreased, boosting the immunological cellular adhesion, despite the increased limitation, on oxygen permeability, expected to impede its utilisation in medical contexts [17]. Some typical types of engineered polymers are discussed below.

PET is a semi-crystalline substrate. Owing to its exceptional mechanical robustness (e.g. a high Young’s Modulus between 2800 and 3100 MPa), excellent visible light transmission, and resistance to either oxidative or moisture deterioration, PET finds versatile applications in flexible electronics. Among all, flexible displays and wearable sensors are typical applications for this material. Polyimide (PI) is characterized by the imide group in the monomers. These monomers could be joined together via the heterocyclic aromatic conjugation [18], which accounts for the extra polymeric structural rigidity and chemical resistance towards common solvents (e.g.oils and esters). Their thermosetting properties, flexibilities, and mechanical strength are retained under temperatures above 230 °C and above 700 °C for short exposure. The material is frequently employed in the production of medical sensors and flexible printed circuit boards (FPCBs). PDMS is a silicon-based organic material known for its biocompatibility, lack of toxicity, and a rubber-like elastic solid nature under low temperatures. Its high permeability to water and oxygen makes it a non-ideal material for OLED, but the production of implantable and wearable devices for medical organ treatments.

3.1.2 Engineering Composites

Composite materials are produced by chemically combining together two or more distinct types of materials in order to obtain a new substance that possesses synergistic characteristics that outperform those of its constituents. Composite materials are of paramount importance in the realm of flexible devices as they facilitate the attainment of improved flexibility and stretchability.

Carbon-nanotube composites (CNT) combine thin carbon tubes in the nanoscale with either metallic elements or any of the engineering polymers above. This combination exhibits both the outstanding electrical conductance from the conductive polymers and the high tensile strength from the carbon nanotube. These composite materials are utilised in flexible electrodes and conductive constituents integrated into flexible devices.

Polymer-ceramic composites incorporate ceramic into polymer matrices, which improves the thermal and mechanical characteristics of the resulting composite, including fracture toughness, strength maintenance at high temperatures, and resistance against environmental corrosion [19]. Flexible circuit boards and electronic components necessitating enhanced durability frequently encompass these composites.

Graphene polymer composites integrate a thin sheet of graphene into the host polymer matrix enhancing the material’s overall mechanical strength (Young’s modulus of 1 TPa and ultimate strength of 130 GPa), chemical and electrical conductivity, and characteristic gas impermeability. It is commonly utilized in flexible display screens as well as sensors where the gas barriers of the host polymer matter.

3.1.3 Device Architecture

Modern scientific advancements have developed multiple ways of integrating bendable and stretchable electrical components.

One method entails coating a flexible and lightweight non-polymeric substrate with the polymers, to create a composite-like complex that combines the chemical rigidity of the engineering polymers with the elasticity and stretchability of the substrate. Yuan et al. have introduced the notion of a paper-based electrode coated with polypyrrole (PPy) through the utilisation of a simple "soak and polymerization" technique (Figure 1). The sustained increase in current indicates that the electrical conductance of the paper electrode remained intact despite undergoing 100 cycles of bending [20].

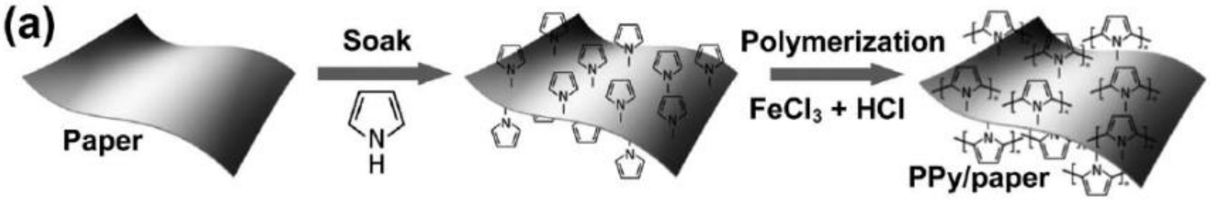

Fig. 1 Soak and Polymerization process for the preparation of PPy [20]

3D integration, on the other hand, refers to the process of vertically stacking (possibly interconnecting) and integrating numerous layers of electronic components, including sensors, transistors, and interconnects. This method improves device performance, allows for miniaturisation, and enhances integration density in comparison to traditional 2D integrated devices. On the other hand, this design distributes the localized mechanical or thermal stress the structure suffers and allows for multifunctionality (more than Moore) [21].

Other architecture strategies for flexible electronics include the encapsulation of target flexible electronics with either organic or inorganic materials, to preserve them from external environmental contamination, even though to some extent restricting the stretchability of the electronics contained. For example, for the thermal sensors, PI, introduced above, stable at 350 °C, could both be utilized as the substrate for the electronics substrate of the sensor or the encapsulation layer protecting the electronics, since this design enables the incorporation via thin film decomposition of e.g. semi-conductor components for the sensor [22].

3.2. Integration Techniques in Wearable Devices

3.2.1 Textile Integration

Recently scientists have developed advanced techniques for embedding organic electronic materials into fabrics:

Firstly, through fabricating a relatively thin layer of organic electronics material onto the fabric substrate, coating processes aim to fulfil the substrate compatibility while preserving the conductivity of the electronics installed. Relevant techniques include spray coating (evenly scattering of material droplets on the substrate, e.g. fabrication of open-sound-control on a textile) [23], homogeneous dip coating for energy harvesting devices e.g. DSSCs [24], and the cost-effective spin coating employing injection and centrifugation which allow even distribution across the substrate surface.

Additionally, unlike the Coating process, Printing treats the substrate as a ‘book page’ and focuses on locating the ‘pattern’ of functional electronics onto the substrate [25], This technique achieves the enhancement in elasticity and stretchability for the integration. Two common techniques are screen printing (transferring ink through a designated mesh or stencil onto a substrate, achieving various applications including sensors) and inkjet printing (propelling ink through a piezoelectric nozzle on the substrate, offering high resolution and cost-effectiveness for short-scale printing) [26].

3.2.2 Skin-Like Electronics

Developments of ultra-skin and electronic skin mimicking skin properties have soared recently in the wearable electronics fields including health monitoring and prosthetic treatments. While providing analytical scanning on diverse health indices, they exhibit extraordinary biocompatibility and are free of external toxicity towards the user skins. The ultra-thin e-skin structure has been demonstrated by combining conductive polymer electrodes of reduced thickness, with high visible light transmission, with elastomer matrices. Liu et.al reported that the lock of chemical bonding in between enables a faster rate of self-healing, transparency as well as the original mechanical flexibility and strength also enables a diverse range of applications for engineering the elastomer [27]. The electrical conductivity is retained after tens of thousands of bending tests, under the voltage generated by frequent human body motion, proving the promising potential in human-machine interactions and conformable skin electronics. However, the challenges are yet to be resolved with regard to different types of skin with various standards of biocompatibility, and the prevention of immunologic or allergic reactions from the human skin under various climates.

4. Applications in Wearable Technology

Wearable technology has witnessed a transformative evolution, with electronics in flexible devices playing a pivotal role in shaping the landscape of applications. This section explores the diverse and impactful applications of flexible electronics in wearable technology, focusing on health monitoring devices, smart clothing, wearable displays, and the power mechanisms driving these innovations.

4.1. Health Monitoring Devices

One of the most significant contributions of flexible electronics to wearable technology is evident in health monitoring devices. The integration of flexible sensors enables the development of devices that can seamlessly conform to the body, allowing for non-intrusive vital signs and movement monitoring. This application has revolutionized personalized healthcare, providing users and healthcare professionals with real-time, accurate data. From heart rate monitoring to tracking physical activity, flexible sensors enhance the functionality and comfort of health-centric wearables.

4.2. Smart Clothing

The integration of electronic components with traditional fabrics is commonly referred to as e-textiles, alternatively known as smart or intelligent garments. The distinctive features exhibited by intelligent garments vary depending on the specific electronic components or technology platforms incorporated within them [28]. The integration of electronic components gives conventional clothing a new functionality that improves its uses and adds value for advanced state-of-the-art applications [28]. The electronic feedback process or functioning mechanism of e-textiles is based on evoking or stimulation received by the senses or any physical response/action from any object or environment in the form of touch, heat, sound, and light [28]. To deliver a response or action, smart e-textiles detect the stimulation and process the information in the form of quantitative data [28]. This technology paves the way for a functional and technologically advanced fashion new era [28]. The Picture below illustrates a pathway of applications of e-textiles in several fields for the prevention of respiratory illnesses, joint injury, skin diseases, neonatal jaundice, road safety, training, and fashion (Figure 2).

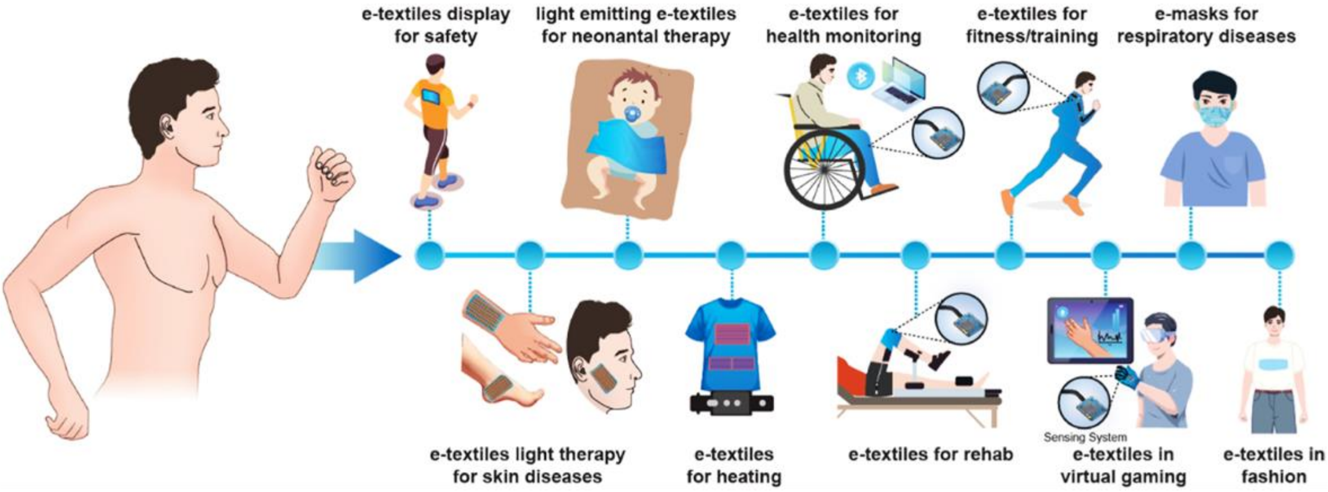

Fig. 2 The pathway of applications of textiles in multiple fields [29].

4.3. Wearable Displays

The development of wearable displays is another area where flexible electronics demonstrate their versatility. Flexible and stretchable screens, crafted using organic materials, offer a novel approach to user interfaces in wearable devices. These displays can conform to irregular surfaces and adapt to various form factors, providing an immersive and dynamic user experience.

To make a device flexible, the key is to use ultrathin materials, especially for the TFT backplane. Thinner materials provide more flexibility. UNIST researchers achieved this by creating a stretchable and transparent backplane using oxide semiconductor TFTs and graphene-Au nanotube (AuNT) hybrid electrodes [30]. These electrodes were highly flexible, and transparent, and maintained good electrical performance even when stretched up to 50% [30]. Another example is a flexible display by Javey and colleagues, featuring a thin CNT-based backplane and flexible OLED pixels [31]. The entire device, excluding the substrate, was less than 2 μm thick, ensuring stable electrical characteristics during bending [31]. This flexible display demonstrated the devices' ability to maintain functionality even when bent [31].

4.4. Powering Flexible Devices: Organic Photovoltaics (OPVs), Flexible Batteries, and Supercapacitors

Ensuring the sustainability and autonomy of wearable devices requires efficient power sources. OPVs, flexible batteries, and supercapacitors emerge as solutions to this challenge.

The inherent flexibility, low weight, and large area processibility of ultrathin (<3 μm) flexible OPVs have garnered significant interest [32]. Although the present power-conversion efficiency (PCE) of flexible OPVs is 17.52%, it is constantly growing [32]. Enhancing the mechanical, operational, and environmental stability of OPVs has been the subject of several studies to facilitate practical integration in wearables skin and tissue-compatible biomonitoring sensors and Internet-of-Things devices that do not require external power sources [33]. Consequently, research on ultra-flexible OPVs has accelerated within the past decade [33].

5. Challenges and Future Perspectives

Despite the promising developments, challenges persist in the widespread adoption of electronics as flexible devices.

Firstly, ensuring the longevity and robustness of flexible devices remains a critical challenge. Organic materials, while contributing to flexibility, often face hurdles in mechanical and environmental stability. The continuous bending and stretching of wearable devices can lead to material fatigue and wear. Moreover, exposure to various environmental factors, such as moisture and temperature fluctuations, poses a threat to the structural integrity of organic components.

Researchers are delving into advanced materials and protective coatings to enhance the durability and stability of organic electronics. Studies, such as the work to explore novel approaches to mitigate mechanical wear and environmental degradation. Developing materials with improved resistance to these stressors is essential for creating wearables that withstand the rigors of everyday use.

Additionally, while promising prototypes exist, transitioning from small-scale production to large-scale manufacturing poses significant challenges. The intricacies of producing flexible devices on a mass scale demand precise manufacturing processes, cost-effective materials, and efficient assembly techniques. Achieving consistency and quality across a high volume of units without compromising flexibility is a complex task.

6. Conclusion

Organic electronics have sparked a significant technological revolution in the dynamic field of flexible and wearable devices, completely changing the way that electronic devices are designed and operated. The crucial function these materials play has been revealed by the investigation of conductive polymers, small molecule semiconductors, and the incorporation of organic dyes and pigments. The foundation for scalable production is laid by synthesis techniques like polymerization and vapor deposition, which provide insight into the direction of large-scale manufacturing in the future.

The emphasis on engineering flexibility and stretchability, coupled with innovative integration techniques such as 3D integration, demonstrates the adaptability of organic electronics in wearable devices. Flexible device design ideas have progressed, from engineering polymers like PET and PI to composite materials like carbon nanotube composites and graphene polymer composites. These developments extend to device architectures that combine chemical rigidity with elasticity, promising a future in which electronics smoothly fit the human body.

The applications in wearable technology underscore the transformative potential of flexible electronics. Health monitoring devices with flexible sensors revolutionize individualized healthcare, while smart clothing incorporates e-textiles, combining fashion and practicality. Wearable displays made of organic materials offer engaging user experiences while challenging traditional electrical interface norms. However, the challenges still exist, with stability and durability leading the way. Researchers are using novel materials and protective coatings to overcome these issues. Attention needs to be given as well to scalability and manufacturing issues in order to achieve widespread commercialization of flexible devices. When considering the advancements in organic electronics, the future holds immense promise. The continual search for improved durability, manufacturing efficiency, and wider applications is a path toward seamlessly integrating flexible electronics into our daily lives. This journey reflects not only technological evolution but also a paradigm shifts in how we perceive and interact with electronic devices, bringing in a new era of wearable and flexible technology.

Authors Contribution

All the authors contributed equally, and their names were listed in alphabetical order.

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