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Toward ultraflexible organic electronic devices

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In recent decades, organic electronics have progressed remarkably, owing to their exceptional mechanical and electrical properties. The development of ultraflexible organic electronics has opened up possibilities for applications such as wearable electronics, flexible displays, and skin- and tissue-compatible sensors with good breathability. In this article, we introduce state-of-the-art ultrathin and high-performance functional organic electronic devices and their integration with potential applications. Our focus is on the strategies for advancing new materials, novel device structures, and diverse applications, which have enabled the development of multifunctional and complex systems. Additionally, we highlight the challenges and opportunities for future research in this field, including improving stability and reliability over long periods, ensuring scalability and cost-effectiveness of production, and addressing issues related to integration into larger systems. Finally, we present future directions in this exciting and rapidly growing field of ultraflexible organic electronics.

Introduction

Over the past few decades, organic electronics have been intensely explored,[1–18] driven by their unique properties that make them highly desirable for a wide range of applications.[19–27] These materials are typically composed of carbon-based molecules that are either (semi-)conducting or insulating,[28–31] allowing a wide range of applications, including transistors,[27,32,33] light-emitting diodes,[34–37] solar cells,[18,25,38,39] and sensors.[40,41] One of the most significant advantages of organic electronics is their mechanical flexibility.[42,43] Unlike the traditional silicon-based devices that are rigid and brittle, organic electronics, typically achieved through the use of polymers, can be made to be extremely flexible and even stretchable[27,44] (**Table I**). This property opens a wide range of possibilities for creation of electronic devices that can conform to different shapes and surfaces,[45,46] including the human body.[42,47]

The development of ultraflexible organic electronics has been a major focus of research, because these devices have the potential to enable a wide range of new applications.[30,48] For example, ultrathin organic thin-film transistors (OTFTs) and their circuits could be used to monitor various biological processes,[27,49,50] including heart rate[41,48,51] and brain activity.[49,52,53] In addition, a recent study has demonstrated the use of organic light-emitting diodes (OLEDs) and photonic systems in flexible and wearable displays[37,54] and sensors,[54–56] including those that can be worn directly on the skin.[41,57] Similarly, organic photovoltaics (OPV) that can be integrated into functional electronic devices for powering wearable devices with accurate sensitivity have been developed.[51,58]

Despite the progress made, several challenges remain. One of the most significant challenges is improving the stability and reliability of these devices over long periods, as organic materials are prone to degradation over time, especially when exposed to environmental factors such as oxygen and moisture. Additionally, these devices require complex manufacturing processes, and finding ways to streamline these processes and reduce costs is crucial. Finally, integrating these devices into complex and larger systems requires new engineering approaches for interconnection.

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Table I. Comparison between conventional and ultraflexible devices.

Aspect	Conventional Devices	Ultraflexible Devices
Form Factor	Rigid and inflexible	Bendable and foldable
Weight	Generally heavier	Lightweight and portable
Flexibility (bending radius, R)	Limited flexibility ($R = \infty$)	Highly flexible ($R < 10$ μm)
Durability	Generally robust	Susceptible to damage
Manufacturing cost	Relatively lower cost	Higher cost due to complex fabrication step
Current Technology Status	Established and mature	Still advancing and evolving
Potential Applications	Traditional use cases	Wearable electronics, unique applications

This article presents the latest advancements in ultrathin and high-performance organic electronic devices, including OTFTs, OLEDs, and OPV, along with their integration into potential applications. By emphasizing the selection of soft materials, the design of innovative device structures, and the diverse range of applications, this article highlights the development of multifunctional systems. The progress achieved in this field has facilitated the creation of ultraflexible electronic devices that can be worn on the skin, seamlessly integrated into clothing, and utilized in various applications.

Furthermore, we discuss the challenges and opportunities for future research in the realm of ultraflexible organic electronics. These challenges encompass enhancing long-term stability and reliability, scaling up production while reducing costs, and resolving issues associated with integrating these devices into complex systems. We also offer insights into the future directions of this rapidly evolving field. We firmly believe that interdisciplinary research and collaboration between academia and industry will play a pivotal role in addressing these challenges and fully unlocking the potential of these remarkable materials. Sustained innovation in ultraflexible organic electronics will open up avenues for groundbreaking technologies and enable a wide range of applications that were once deemed unattainable.

Materials and structures of ultrathin organic electronics

In order to realize ultrathin and high-performance organic electronic devices (**Figure 1**a), three primary strategies have been proposed, each requiring specific elements to achieve mechanical flexibility and fundamental knowledge concerning materials properties and integrated device architecture (Figure 1b). These strategies are as follows: (1) Careful selection of soft materials with high yield strengths (Figure 1c) is a key approach to be considered. (2) Geometric engineering of the electronic devices (Figure 1d) represents another crucial strategy to pursue. (3) Appropriate passivation to position the devices on a neutral plane (Figure 1e) serves as an important aspect to address. The neutral plane denotes a specific location within the device structure that remains unaffected or experiences minimal strain during bending or deformation.[59,60] This region serves as a crucial reference plane, dividing the device layers into two distinct zones: the compression zone, which encounters compressive stress, and the tension zone, subject to tensile stress during bending or stretching. Acquiring a clear understanding of the neutral plane's location is vital for effectively designing and optimizing the mechanical performance of these devices.

The selection of soft materials is pivotal for the progression of ultrathin electronic devices, primarily due to their ability to withstand the mechanical deformations experienced during regular device use (Figure 1c). These materials are key to the development of electronics that are simultaneously ultrathin and flexible, given that they can deform without damaging their mechanical or electrical properties. Soft materials characterized by a low Young's modulus can endure deformations, avoiding fracture and maintaining their electrical characteristics, thereby ensuring the device's functionality and longevity. The capacity of these materials to absorb and disperse mechanical stresses within their matrix prevents mechanical failure, while their ability to maintain their properties even under significant deformations contributes to the devices' durability and reliability. The choice of soft materials not only influences the device's mechanical flexibility, conformability, stability, and durability, but also its electrical performance. Depending on their specific characteristics, these materials can exhibit varying levels of conductivity, semiconductivity, or insulation. For semiconductors and conductors, the soft materials' stress relaxation ability, charge transportation properties, and resistance to fatigue are pivotal factors that significantly influence the device's overall performance and lifespan. On the other hand, for dielectrics, maintaining a condensed structure becomes crucial to minimize leakage current and capacitance under mechanical deformation. This ensures optimal performance and reliability for devices that rely on dielectric materials. Given the potential damage severe deformations can cause, such as mechanical fatigue and device failure, it is crucial to select the right soft materials to preserve the operational stability of ultrathin electronic devices. As these devices increasingly permeate various sectors, the importance of suitable soft material selection will undoubtedly grow, thereby magnifying their role in optimizing device performance across diverse fields.

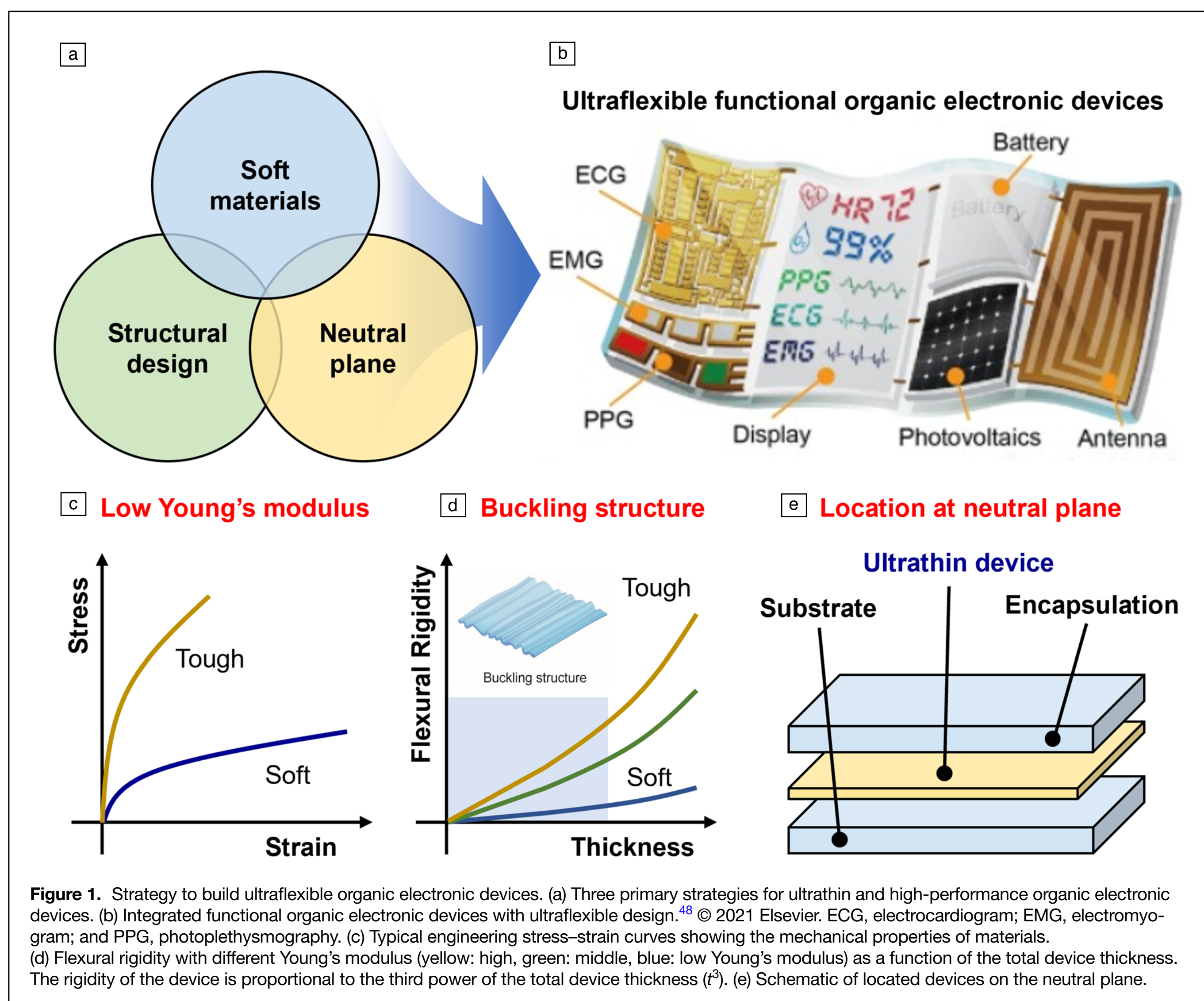

Figure 1. Strategy to build ultraflexible organic electronic devices. (a) Three primary strategies for ultrathin and high-performance organic electronic devices. (b) Integrated functional organic electronic devices with ultraflexible design.[48] © 2021 Elsevier. ECG, electrocardiogram; EMG, electromyogram; and PPG, photoplethysmography. (c) Typical engineering stress–strain curves showing the mechanical properties of materials. (d) Flexural rigidity with different Young's modulus (yellow: high, green: middle, blue: low Young's modulus) as a function of the total device thickness. The rigidity of the device is proportional to the third power of the total device thickness (t^3). (e) Schematic of located devices on the neutral plane.

The geometric structure significantly impacts the flexural rigidity of ultrathin electronic devices, playing a vital role in their design (Figure 1d). The design and overall structure directly affect the mechanical attributes of the device, including but not limited to mechanical flexibility, stiffness, and strength. The strategic layout of this structure also influences the distribution of stress within the device, thereby lowering the potential for mechanical failure. Flexural rigidity, indicative of a material's resistance to bending, is a crucial element in the design of ultrathin electronic devices. A well-crafted geometric structure can effectively reduce flexural rigidity, leading to better integration with other components and facilitating conformal adhesion. Geometric designs, including serpentine patterns, island-bridges, and strain localization methods, are crucial for accommodating mechanical stress and strain in flexible devices.[61] These designs allow the device to flex and conform to various shapes without compromising its structural integrity. By incorporating island-bridges, stretchability is enhanced while maintaining overall rigidity. Moreover, strain is directed or concentrated to specific regions, resulting in a notable improvement in the device's overall mechanical behavior.

Aspects of the geometric structure such as the overall thickness of the devices and the choice of materials with a low Young's modulus contribute to the minimization of flexural rigidity. This reduction allows the creation of ultraflexible devices featuring a buckling structure, which forms a pattern of waves or wrinkles when subject to compressive loads. Buckling structures not only accommodate device deformation without breakage, thereby enhancing mechanical stability and reliability, but also increase the stretchability of ultrathin electronic devices. These buckling waves or wrinkles absorb the tensile stresses within the device, enabling it to stretch without the risk of rupture. This attribute is particularly advantageous in applications where the device must conform to curved or irregular surfaces.

Strategically positioning ultraflexible organic electronic devices in a neutral plane, coupled with effective passivation, is key to maintaining their functionality and reliability (Figure 1e). Locating the device in a neutral plane and ensuring thorough passivation can lessen the mechanical stress experienced by the device, thereby enhancing its mechanical stability and reliability. The neutral plane refers to a plane where the material endures zero stress when exposed to mechanical deformation. By positioning the device within this plane, we can minimize mechanical stress, reducing the mechanical failure. This placement strategy is particularly crucial for ultraflexible organic electronic devices, which undergo extensive deformations during operation. Furthermore, effective passivation shields the device from environmental elements such as moisture, oxygen, and UV radiation, which can degrade its performance. Proper passivation also improves the device's electrical properties by diminishing the occurrence of defects or impurities that can hinder its performance.

For the successful realization of ultrathin, high-performance organic electronic devices, it's crucial to carefully introduce soft materials with high yield strengths. This ensures mechanical flexibility and stability within the device. Furthermore, the geometric design of these devices should aim to minimize their flexural rigidity, facilitating the creation of a buckling structure adding stretchability. An equally important factor is the strategic passivation and positioning of the devices within a neutral plane. This not only guards the device against mechanical strain and environmental factors that could degrade performance, but also helps maintain consistent electrical properties. Soft organic materials vary greatly, each with unique properties and uses. Mechanical materials such as elastomers and gels exhibit significant deformation under stress, used in seals, medical applications, and cosmetics. Electrically, conductive polymers allow current flow, used in OLEDs and antistatic substances, whereas dielectric polymers, like polyethylene, serve in capacitors and insulation. Optically, OLED materials emit light when electrified, used in digital displays, and photochromic materials change color under light, used in eyeglasses and plastics. Thermally, insulation materials slow heat transfer, used in buildings and packaging, whereas thermoset polymers harden upon heating, used in coatings and high-strength parts. These categories aren't exclusive, with many materials exhibiting properties from multiple categories. Employing these strategies facilitates the development of ultraflexible, reliable organic electronic devices, offering invaluable insights into the core mechanical properties of materials and device architecture. This knowledge has the potential to guide future research in this field. By pursuing these strategies, ultraflexible functional organic electronic devices and their practical applications, including health monitoring, medical therapy, and soft robotics are reviewed.

Ultrathin organic thin-film transistors

Organic transistors offer the possibility of fabricating flexible devices owing to the wide substrate selectivity in the manufacturing process and the inherent flexibility of organic materials. Particularly, the stress imposed when bending can be dramatically reduced by decreasing the total thickness of the device through the use of thin substrates. This makes it feasible to manufacture highly flexible transistors and their circuits. In this section, we discuss the recent progress on ultrathin transistors and their circuits with a thickness of 10 μm or less.

For ultrathin OTFTs, handling techniques during the fabrication process are crucial because the thin substrates are prone to deformation. Transferring thin films onto rigid and thick substrates[42] or forming thin films with sacrificial layers on thick substrates[27,62,63] has been proposed, where the OTFTs on thin films can be delaminated after the fabrication process. For example, ultrathin OTFTs could be manufactured on the ultrathin substrate by transferring a 1.2-μm-thick poly(ethylene naphthalate) (PEN) film to poly(dimethylsiloxane)-(PDMS)-coated thick PET film.[42] An overall thickness is approximately 3 μm and the resulting transistors are extremely lightweight (less than 3 g/m^2). Additionally, leveraging its super mechanical flexibility makes it possible to fabricate effectively stretchable devices, although the OTFTs are fabricated on nonstretchable plastic substrates. When attaching the ultrathin devices to prestretched rubber and then releasing the rubber, a wrinkled structure is formed with a bending radius ranging from a few to tens of micrometers. Nevertheless, the excellent bending durability of the transistors maintains their original performances despite the formation of such wrinkles.

Considerable approaches have been introduced to improve the electrical/mechanical properties of ultrathin OTFTs or realize other functionalities. The high-quality semiconducting layers on ultrathin OTFTs can be obtained through methods such as crystal engineering,[33,64] interface engineering,[65,66] and transfer processes.[67,68] For example, highly crystalline C10-DNTT is deposited on a separate substrate, covered with a protection layer followed by its transfer to the target substrate through a solution process.[67] This approach could enable achieving high mobility (5.1 cm/Vs) and high bias-stability. Compatibility with the printing method is also important for fabricating large-area OTFTs, which provides the capability of a roll-to-roll process. Indeed, fully printed OTFTs were reported and their layers were formed on the 1-μm-thick parylene substrate by using printing methods such as spin coating, inkjet printing, dispensing, and so on.[69] Reducing the overall thickness of OTFTs makes it possible to introduce new functions such as self-adhesiveness[70] and higher mechanical flexibility. Transistors with a thickness of approximately 300 nm were developed[71] and formed on a few micrometer-thick water-soluble polymer (poly(vinyl alcohol) (PVA))

layer. The PVA layer enabled the handling of submicron-thick OTFTs; then it was removed by applying water. The OTFTs could be adhered to the skin without using gel or other adhesives. Recently, a solution-assisted delamination method was reported to realize the fabrication of sub-150-nm-thick OTFTs (**Figure 2**a).[32] The poly(vinyl formal) (PVF) layer with only few tens of nanometers was formed on a poly(diallyldimethylammonium chloride) (PDAC)/Si wafer. When the PVF/PDAC/Si wafer was immersed into the water, the freestanding PVF layer floated on the water and spread out due to its high hydrophobicity. Finally, by attaching two PVF layers (one layer with semiconductor and source/drain electrodes, and the other layer with gate electrodes), the total thickness of OTFTs could reach below 150 nm. Another important approach was to use porous substrates to achieve new functions that could not be achieved with continuous films. Particularly, introducing electrospun nanofibrous substrates has been proposed, where the semiconductors are thermally evaporated[72] or spray-coated[73] onto the substrates (Figure 2b). The nanomesh structures allow high bending durability resulting from the deformation of the fibers in response to external stress. Furthermore, their porous structures result in excellent gas/mass permeability. This is important in situations where the mass interactions between the OTFT-attached object and to the outside are required.[74,75]

The ultraflexiblity of ultrathin OTFTs has significantly expanded the adaptability of the electronics to various fields, such as wearable,[76,77] implantable,[24] plant,[78] and soft robotic[79] applications. They can be applied to soft objects with curved surfaces, and effectively reduce changes in mechanical properties because of the addition of electrical functions to the objects. For example, an ultrathin OTFT active matrix (12 × 12) combined with a nanofibers-based pressure sensor was attached to the soft elastomer and the detection of the pressure distribution could be successfully demonstrated (Figure 2c). The biological tissues/organs are also important objects. The intimate and conformal contact between the

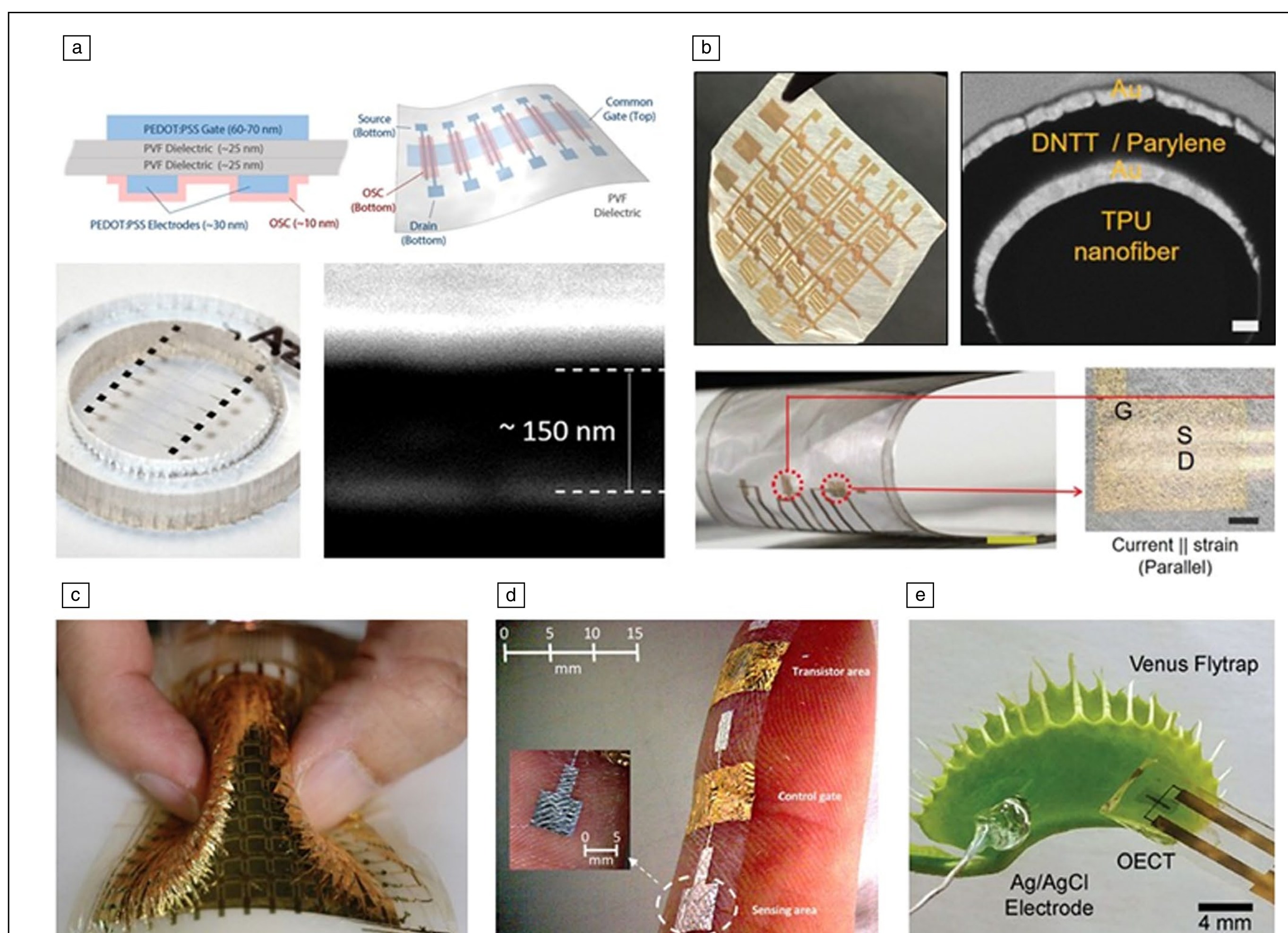

Figure 2. Ultrathin organic thin-film transistors (OTFTs). (a) Sub-150-nm OTFTs using solution-assisted delamination process.[32] © 2021 Nature Publishing Group. (b) Nanomesh OTFTs fabricated on electrospun nanofibrous substrates.[72] © 2022 Wiley. (c) Ultrathin 12 × 12 OTFTs active matrix array on soft elastomer.[79] © 2016 Nature Publishing Group. (d) Ultrathin OTFTs applied to the skin.[77] © 2018 Nature Publishing Group. (e) Organic electrochemical transistor (OECT) on Venus flytrap.[78] © 2020 Wiley.

electronics and the biological objects can be achieved by using ultrathin OTFTs. Biological signals can be acquired at the signal sources or further amplified *in situ* (Figure 2d–e). It would help to suppress artifacts caused by contact instability due to motion and contamination of noises in the wiring.

Ultrathin organic optoelectronic diodes

OLEDs have been extensively developed for display applications owing to their advantages such as vivid colors and wide viewing angles.[80,81] The significant challenges in the development of OLEDs on ultrathin substrates involve indium tin oxide (ITO) electrodes, which are transparent and conductive but not shows good mechanical flexibility, and the air-stability of the devices. To address the problem of transparent electrodes using poly(3,4-ethylenedioxythiophene):polystyrene sulfonate (PEDOT:PSS) instead of ITO, the first ultraflexible OLED has been realized.[82] The ultraflexible OLEDs, which were fabricated on a 1.2-μm-thick poly(ethylene naphthalate) (PEN) film substrate, exhibited high mechanical flexibility, withstanding bending and twisting without breaking. However, these ultraflexible OLEDs had a lower external quantum efficiency (EQE) than 0.5% and could only operate in a nitrogen atmosphere. Overcoming these problems, photonic skins have been reported that can be operated in air and have an EQE of more than 10 percent.[41] To achieve operation in air, these ultraflexible OLEDs have ITO electrodes as transparent electrodes and a stacked film of inorganic and organic materials as an encapsulation layer. To enhance the mechanical durability of the OLEDs, other groups used silver nanowire electrodes as transparent electrodes instead of ITO electrodes.[83] Furthermore, these ultraflexible OLEDs could be combined with prestretched elastomer substrates to realize stretchable OLEDs.[84] Owing to the development of the fabrication process and materials, the currently ultraflexible OLED display was realized by combining MoS_2 TFT active matrix and Green OLEDs[37] (**Figure 3**a).

However, long-term stability of ultraflexible OLEDs remains a challenge for sensor applications. There are mainly two approaches that can be considered to address this challenge: (1) developing an ultrathin passivation film with good mechanical flexibility and high gas barrier properties and (2) improving the stability of the OLED itself. In general, a stacked structure of organic and inorganic layers was used to realize an ultrathin passivation film with mechanical flexibility and highly gas barrier properties. For example, Al_2O_3 and a polymer material, including S–H nanocomposite were used in the inorganic and organic layer, respectively, and showed high gas barrier properties of 1.76×10^5 gm^{-2} after being bent 100 times at a curvature radius of 3 cm, despite having a thickness of less than 1 μm in seven layers.[85] Furthermore, by using Al_2O_3/MgO (AM) nanolaminates as the inorganic layer, a flexible OLED that operates stably without dark spots for 50 days at 60°C and 90% relative humidity has been reported.[86] One of the most effective approaches is the use of inorganic layers as barrier films. Particularly, Al_2O_3/MgO (AM) nanolaminates have been reported to enable stable operation of flexible OLEDs without any dark spots under 60°C, 90% relative humidity for up to 50 days. Moreover, the stability of the OLED devices has significantly improved in recent years, and one of the key factors behind this is the use of an inverted structure.[87] In conventional OLED structures, an ITO electrode and an air unstable material such as Ca or LiF/Al are used as the anode and cathode, respectively.[35,36,88] Therefore, a high-barrier passivation layer is required for stable operation in the air. In contrast, the inverted structure uses an ITO electrode as the cathode and ZnO/polyethylenimine (PEI) as the electron injection layer on top of the ITO electrode, which enables both high stability and high efficiency (Figure 3b). These approaches have also been applied to ultraflexible OLEDs, resulting in OLEDs that can operate underwater[34]or in the air for over 10 days.[57] The achievement of such high stability has enabled the development of novel applications of ultraflexible OLEDs. For instance, OLEDs have been integrated into wound dressings to provide a light source for photodynamic therapy[55]or used as light sources for optogenetic nerve stimulation[53] (Figure 3c).

Ultrathin organic solar cells

The emergence of non-fullerene acceptors represented by the Y-series achieved high power-conversion efficiency (PCE) of organic solar cells. After the PCE of organic solar cells jumped to more than 15% using non-fullerene acceptors first reported in 2019,[89] various additional strategies and materials have led to energy-conversion efficiencies exceeding 19 percent.[90–92] The recent main strategy is to control the morphology of the donor and acceptor domains using a ternary mixture of donor–acceptor blend films to achieve an appropriate balance between improving the efficiency of charge generation and suppressing recombination loss.

Emerging photovoltaics (PV), including organic solar cells, perovskite solar cells, and quantum dot solar cells, have a thin optimized photoactive layer ranging from 100 nm to less than 1 μm, and a unique feature that a flexible polymer film can be adopted as a substrate film, which enables flexible PV devices. With such mechanical flexibility, the emerging PV are expected to generate power in a wide range of places, not limited to rooftops and flat ground as assumed for conventional silicon solar cells. As a solar cell is an energy-harvesting device whose power generation is determined by its area, reducing the device thickness can provide additional benefit of reduced weight (**Figure 4**a).[93,94] It has been shown that an organic solar cell can be stably fabricated on a polymer film with a thickness of down to 1 μm. Usage of such thin film as a substrate realizes a power generation amount per unit weight of more than 10 W/g. With the help of recent improvements in materials for photoactive layers, an excellent PCE of 17.3% was reported with an organic solar cell fabricated on 1.3-μm-thick polyimide substrates, corresponding to 39.7 W g^{-1} as a calculated power per weight.[95]

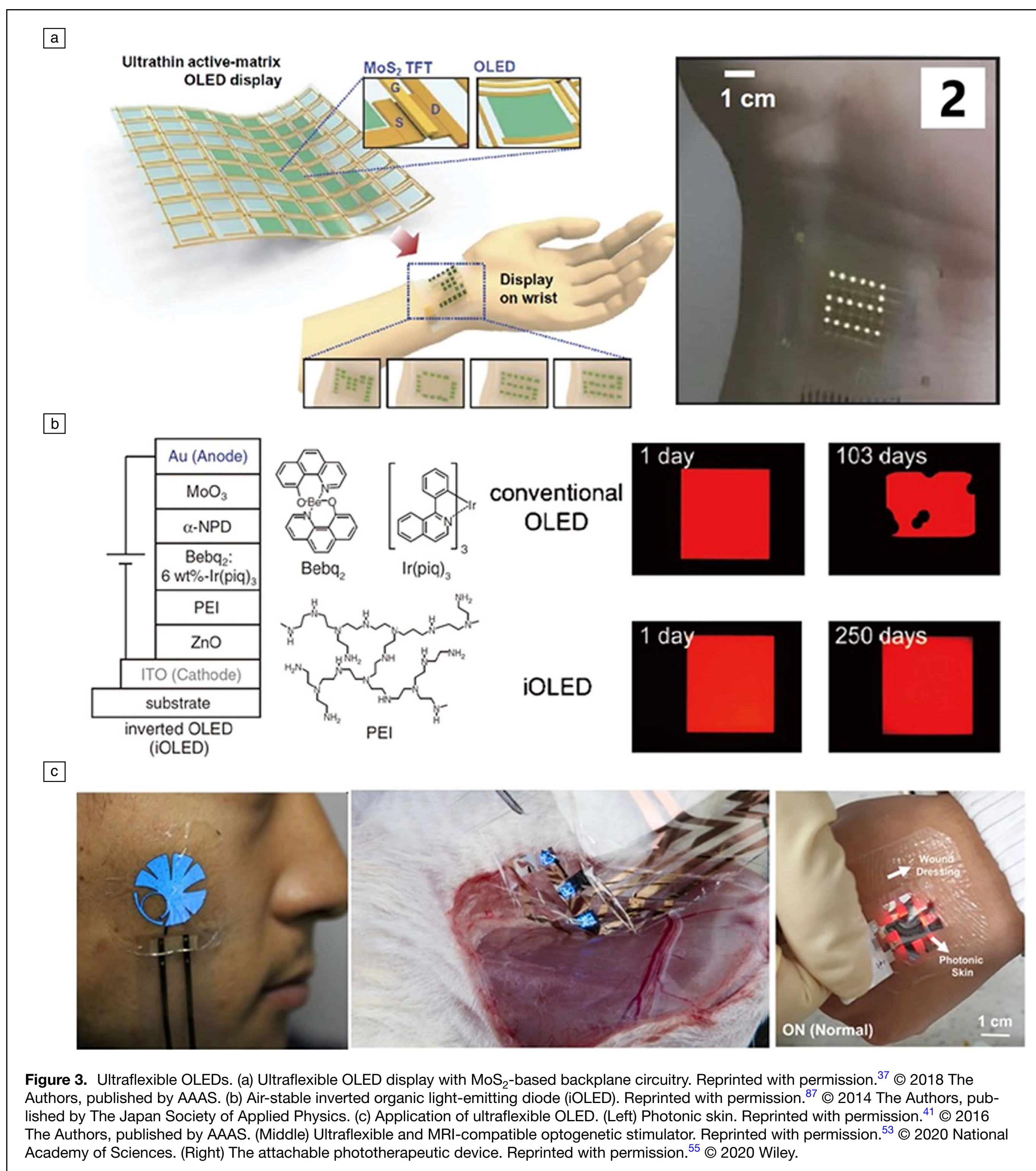

Figure 3. Ultraflexible OLEDs. (a) Ultraflexible OLED display with MoS_2-based backplane circuitry. Reprinted with permission.[37] © 2018 The Authors, published by AAAS. (b) Air-stable inverted organic light-emitting diode (iOLED). Reprinted with permission.[87] © 2014 The Authors, published by The Japan Society of Applied Physics. (c) Application of ultraflexible OLED. (Left) Photonic skin. Reprinted with permission.[41] © 2016 The Authors, published by AAAS. (Middle) Ultraflexible and MRI-compatible optogenetic stimulator. Reprinted with permission.[53] © 2020 National Academy of Sciences. (Right) The attachable phototherapeutic device. Reprinted with permission.[55] © 2020 Wiley.

Such a figure of merit can work effectively for target surfaces that dislike weight. A common potential application is using this organic solar cell as a power source for wearable electronics.[58,95] Such ultrathin organic solar cells can be useful for miniature robotics and soft robotics applications. Ultrathin organic solar cells can generate energy without interfering with the movement of small robots whose weight is directly related to their motion abilities (Figure 4b).[96] Another potential application is energy harvesting in space. The transportation cost to space still exceeds 1000 $/kg;[97] therefore, the reduction of weight can be an important feature for energy harvesting in outer space. For practical applications, solar cells

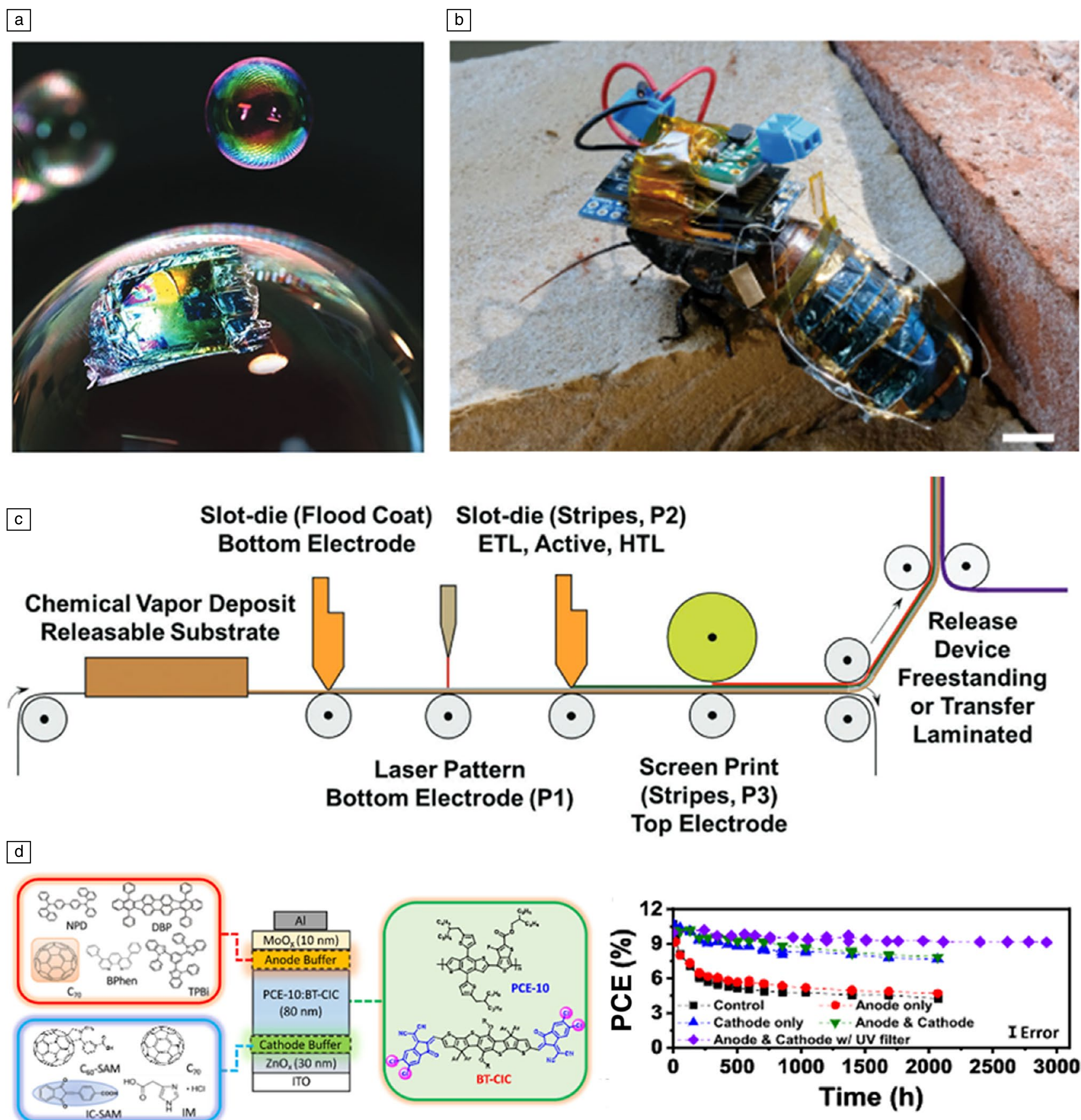

Figure 4. Flexible organic solar cells. (a) Ultrathin organic solar cells on the crown of a dandelion, representing their lightweight property, and (b) potential application for wearable power source. Reprinted with permission from Reference 94. © 2020 Wiley. (b) Ultrathin organic solar cells adhering to living insect bodies that can power a cyborg insect. Reprinted with permission from Reference 96. © 2022 Nature Publishing Group. (c) Schematic illustration of how ultrathin organic solar cells can be manufactured in a scalable manner using printing processes. Reprinted with permission from Reference 98. © 2023 Wiley. (d) Schematic of the device showing layer thicknesses and compositions and its power-conversion efficiency (PCE) change versus aging time under 1 sun simulated AM1.5 G illumination. Reprinted with permission from Reference 102. © 2021 Nature Publishing Group. ETL, electron-transport layer and HTL, hole-transport layer.

combine multiple cells in series or parallel to make a module. It is necessary to consider the design of ultrathin organic solar-cell modules to reduce the margins that do not contribute to PV properties and increase the power generation per weight of the whole module, instead of power per weight of effective area. Additionally, printing processes should be an important key technology to achieve ultrathin organic solar cells in a large area (Figure 4c);[98] therefore, both PCE and stability of printed

organic solar cells should catch up with state-of-the-art organic solar cells with vacuum evaporated layers.

Stability is an important parameter for flexible organic solar cells as well as improved PCE.[99] Recent studies reveal that both donors and acceptors of organic solar cells using non-fullerene acceptors experience degradation from environmental parameters such as photochemical decomposition of Y-series acceptors[100] and backbone twisting of polymers under light irradiation and ambient air.[101] Recent studies report that perfect passivation with glasses and UV-cut filters/LED light that do not include UV wavelength can achieve long-term operation stability of organic solar cells over 10 years with appropriate active layer choice and interfacial improvement (Figure 4d).[18,39,102] In case of ultrathin organic solar cells where it is difficult to ensure sufficient gas barrier properties, it is necessary to consider the balance between setting the device lifetime according to the application and the loss of mechanical flexibility due to the provision of barrier properties. Mechanical flexibility is also an important parameter for flexible organic solar cells. In addition to the brittleness of the functional layer itself,[38,44,103] the interfacial adhesion has been shown to influence the mechanical robustness of the device.[104,105]

Integration strategy of ultraflexible organic electronics for new functionality

As discussed in the previous sections, the performance of flexible electronics with ultrathin and lightweight properties are becoming comparable with those of rigid electronics, such as power-conversion efficiency and EQE. Therefore, the next challenge for fabricating practical next-generation wearable devices is related to the system integration method, retaining ultraflexible and lightweight properties. This section focuses on the bonding method for integrating the electronics and new functionality via device-to-device integration.

The bonding method for flexible electronics is divided into two approaches. The first approach is inserting the adhesive layers between the flexible electronics. The usual adhesive bonding methods are anisotropic conductive adhesive (ACA) (**Figure 5**a)[106] and anisotropic conductive film (ACF).[107,108] ACF/ACA, which contains the conductive filler into an insulating polymer adhesive, develops the conductive pass between electrodes by sandwiching the conductive filler between electrodes under heat (room temperature~200°C) and pressure (2–5 MPa).[109] This method is versatile, and multiple electrodes, such as the anode and cathode, can be bonded simultaneously because of many candidate materials that can be bonded and one-way conductive pass. The current research related to the ACF/ACA could achieve 10 um of high-resolution bonding pitch owing to UV-cured insulating adhesive with reducing conductive filler diameter.[106] Researchers have developed a metal-coated insulating filler[110] and nanofiber/solder ACF,[111] which combined with nanowire and metal particles, achieves excellent mechanical durability owing to the prevention of electrode surface damage under pressure. In addition to ACA/ACF technology, silver paste and silver nano-ink with eutectic gallium indium[112] have developed as a conductive intermediate layer.

The second approach is direct bonding without any adhesive between electrodes. Although conductive adhesive bonding is versatile, the thickness of the adhesive causes a rapid increase in the bending rigidity and lowers the mechanical flexibility of the flexible electronics. In contrast, the direct bonding method is one of the most effective ways to fabricate integrated flexible electronics with keeping the original flexibility because the thickness of the bonding region does not increase by the thickness of the adhesive self. The biphasic, nanodispersed (BIND) bonding method is one of the most practical direct bonding methods (Figure 5b).[113] In BIND bonding, the rubber-based material, styrene-ethylene-butylene-styrene (SEBS), is used as a substrate, and the exposure balance between Au and SEBS on the substrate surface is controlled by optimizing the Au deposition speed and film thickness on SEBS to create a unique BIND state that combines the self-adhesiveness of SEBS with the conductivity of Au. As a result of optimization, BIND bondable wiring is created by depositing Au at a deposition speed of 0.5–1.0 Å/s and a film thickness of 45–60 nm. The direct bonding of the flexible bonding achieved with this special wiring, a conductive path is formed by contact between Au wiring due to self-adhesion between the SEBS substrate. The permissive minimum bonding pitch of the BIND bonding is 100 µm, and it exhibits conductivity even at 180% stretch owing to the stretchability of the SEBS material. Additionally, BIND bonding can apply to bonding not only flexible electronics, but also rigid substrates and soft material bonding owing to the adhesiveness of the SEBS material. In addition to BIND bonding, the water vapor plasma-assisted bonding (WVPAB) method, a direct bonding method by metal bonding of Au electrodes, is effective when low contact resistance and high-resolution bonding are required (Figure 5c).[114] In the WVPAB method, Au wiring evaporated on a polymer substrate is irradiated with water vapor plasma. The treated surfaces contact each other in the air and are left in the air without heating for 12 h or more, resulting in metallic bonding between Au electrodes. The OH groups contained in the water vapor plasma improve the adhesion of the bonding interface and promote surface-activated bonding of the Au electrodes, resulting in direct bonding even between Au electrodes with a rough surface (RMS: around 7 nm). The WVPAB method can achieve low contact resistance of less than 0.1 Ω with a wiring pitch of 10 µm or less owing to metallic bonding between Au electrodes. By reducing the surface roughness of the bonding surface, the WVPAB method can apply to bonding thin-film polymer substrates and chip LEDs. To advance the practical application of wearable electronics systems, it is critical to develop a bonding method that combines high resolution, bonding strength, and low contact resistance. Such a method would enable the creation of wearable electronic devices that can be worn comfortably on the body, conform to different body shapes and movements, and maintain their functionality over time. Therefore, establishing a bonding method

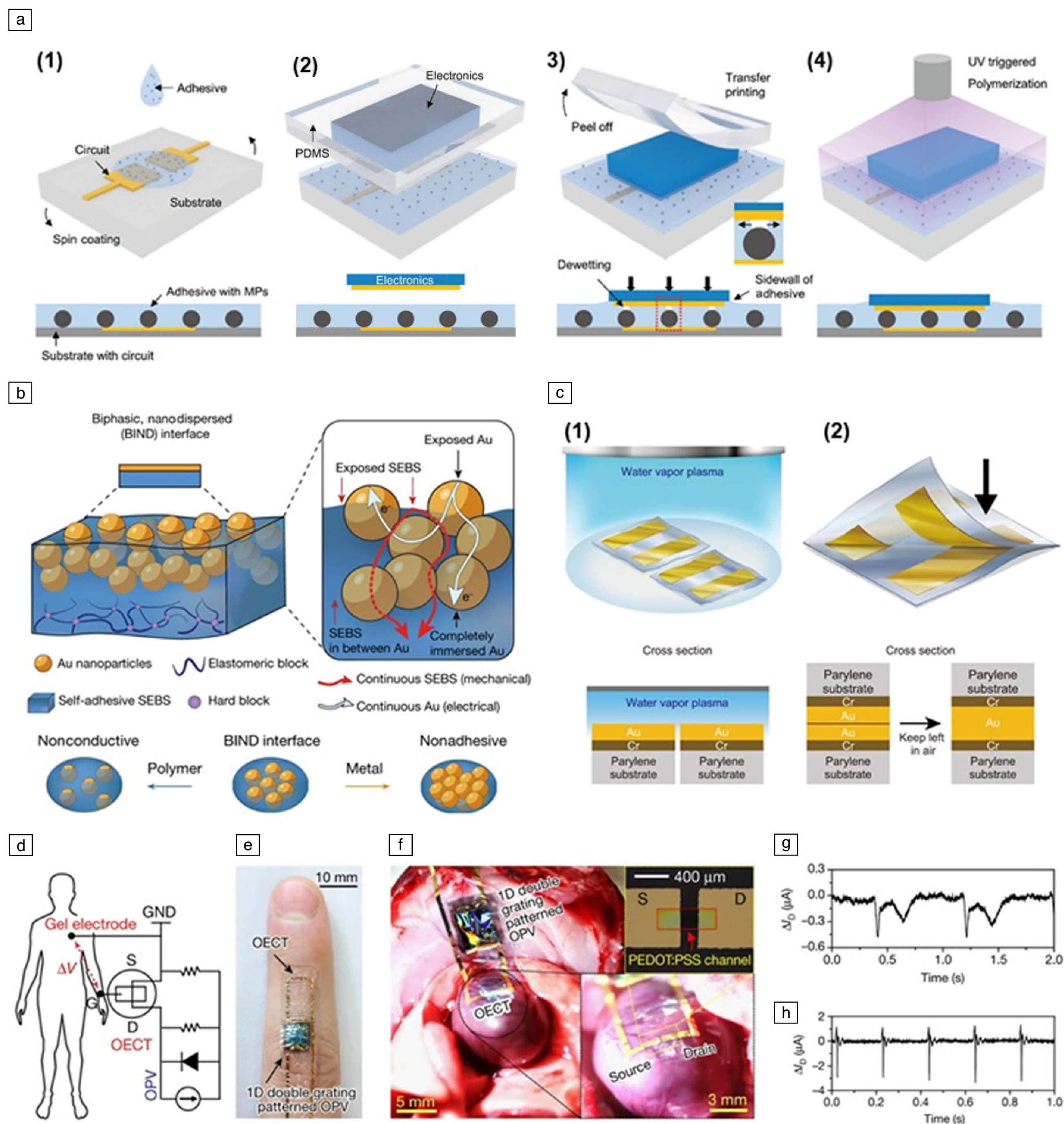

Figure 5. Current bonding methods for integrated flexible electronics. (a) Nanoscale-dewetting-driven anisotropic conductive adhesive (ACA) bonding method.[106] (1) Spin-cast ACA on substrate with metal electrodes. (2) Alignment for electrical interconnection between metal pads of the electronics and the prepatterned metal circuit of the substrate using a PDMS stamp. (3) Deterministic transfer printing of the electronics and interconnection based on nanoscale dewetting of the unstable polymer adhesive (inset). (4) UV flood exposure for the polymerization of the adhesive. © 2020 Wiley. (b) Biphasic, nanodispersed (BIND) bonding method. The BIND bonding requires controlling the balance of SEBS substrate self and Au conductive nanoparticle on bonding region surface by the deposition rate and layer thickness. Larger deposition rates and thickness result in non-bonding, whereas smaller ones render the interface nonconductive. © 2023 Nature Publishing Group.[113] (c) Water vapor plasma-assisted bonding method.[114] (1) The water vapor plasma exposes the Au electrode surfaces fabricated on 2-μm-thick parylene substrate to activate the surface and attach the OH chemical group. (2) Au direct bonding was achieved by contacting the treated surfaces and keeping them in ambient air without heating for more than 12 h.[58] (d) Wiring diagram for cardiac signal recording. The chest and source electrodes of the integrated device were grounded and the potential difference (ΔV) between the chest and active channel of the organic electrochemical transistor (OECT) functioned as the gate bias. (e) Photograph of the self-powered integrated electronic device attached to a finger. (f) Photograph of the self-powered integrated electronic device attached to the heart of a rat (left), and enlarged images of the channel area (right, top) and of the source–drain electrode (right, bottom). Measured output current from the recorded cardiac signal trace from skin (g) and tissue (h) surface under light illumination. (d, e) Reprinted with permission from Reference 58. © 2018 Nature Publishing Group. OPV, organic photovoltaics and GND, ground.

that possesses these characteristics is essential for unlocking the full potential of flexible electronics in wearable applications.

Integration systems are essential in driving the development of next-generation devices, as they enable the creation of sophisticated and multifunctional devices that cater to the diverse and evolving needs of users. The seamless combination of various components and technologies allows these systems to perform multiple tasks efficiently, eliminating the necessity for separate, specialized devices and providing users with a more streamlined and convenient experience. As technology continues to advance at an unprecedented pace, there is an increasing demand for compact, user-friendly, and energy-efficient devices that can perform a wide array of functions. Integration systems address this demand by optimizing the use of available resources, minimizing energy consumption, and maximizing performance across different functionalities.

The importance of integration systems is more apparent in wearable technology, health care, and IoT, where the combination of multiple functionalities into a single device can significantly improve the quality of life for users. Integrated wearable devices have the capability to monitor vital health parameters[56,58] track physical activities,[51,115] and provide personalized feedback,[25,40,116] all while maintaining a comfortable and unobtrusive form factor. In health care, the development of integrated sensor systems allows for continuous and noninvasive monitoring of patients, leading to more accurate diagnoses, timely interventions, and improved patient outcomes. For instance, the self-powered ultraflexible solar cell can be used as part of an integrated sensor system to monitor physiological conditions, enabling real-time monitoring and analysis of health data (Figure 5d–h). Ultimately, integration systems are pivotal in shaping the future of technology, as they push the boundaries of what is possible and redefine our interactions with technology. By fostering innovation and enabling the creation of devices with unprecedented capabilities, integration systems have the potential to revolutionize various industries and improve the quality of life for people around the world.

Conclusion

Organic electronics have demonstrated unique and exceptional properties that make them highly desirable for a wide range of applications. The mechanical flexibility of these materials is one of their most significant advantages because it enables the creation of electronic devices that can conform to different shapes and surfaces, including the human body. The development of ultraflexible organic electronics has been a major focus of research in recent years, with potential applications in data processing, visualization, energy, and sensors with highly integrated and multifunctional systems. However, several challenges remain to be addressed. To achieve long-term stability and reliability, it is crucial that the operational lifetime of these devices is increased by an order of magnitude compared to current standards. Scaling up production and reducing costs also pose significant challenges, with a target performance goal of reducing production costs by at least 50% while increasing yield to 90% or higher. Addressing issues related to the integration of these devices into complex systems is another critical area of focus. Optimal integration would entail seamless inclusion of these devices into systems without compromising their performance. Despite these challenges, we believe that interdisciplinary research and collaboration between academia and industry will be critical to address them and fully realize the potential. Future innovation in ultraflexible organic electronics will pave the way for new technologies, enabling a wide range of applications that were once considered impossible. The potential benefits of these materials are enormous and could revolutionize many industries, from health care to energy. Achieving these benefits necessitates reaching these specified performance targets, which will require continued R&D. Overall, we believe that ultraflexible organic electronics will continue to be a rapidly growing and exciting field with enormous potential for future technological advancements.

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Conflict of interest

On behalf of all authors, the corresponding author states that there is no conflict of interest.

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