coatings

MDPI

Review

Prospects and Challenges of Flexible Stretchable Electrodes for Electronics

Wei Hou [1], Qingwei Liao [1,*], Shuang Xie [2,*], Yujun Song [3,*] and Lei Qin [1,*]

[1] Key Laboratory of Sensors Technology, Beijing Information Science & Technology University, Beijing 100192, China; houwei112@163.com
[2] School of Microelectronics, Shandong University, Jinan 250101, China
[3] Center of Modern Physics Technology, School of Mathematics & Physics, University of Science & Technology, Beijing 100083, China
* Correspondence: liaoqingwei@bistu.edu.cn (Q.L.); shuang.xie@sdu.edu.cn (S.X.); songyj@ustb.edu.cn (Y.S.); qinlei@bistu.edu.cn (L.Q.)

Abstract: The application of flexible electronics in the field of communication has made the transition from rigid physical form to flexible physical form. Flexible electrode technology is the key to the wide application of flexible electronics. However, flexible electrodes will break when large deformation occurs, failing flexible electronics. It restricts the further development of flexible electronic technology. Flexible stretchable electrodes are a hot research topic to solve the problem that flexible electrodes cannot withstand large deformation. Flexible stretchable electrode materials have excellent electrical conductivity, while retaining excellent mechanical properties in case of large deformation. This paper summarizes the research results of flexible stretchable electrodes from three aspects: material, process, and structure, as well as the prospects for future development.

Keywords: flexible stretchable electrodes; flexible electronics; electrode material

Citation: Hou, W.; Liao, Q.; Xie, S.; Song, Y.; Qin, L. Prospects and Challenges of Flexible Stretchable Electrodes for Electronics. *Coatings* **2022**, *12*, 558. https://doi.org/10.3390/coatings12050558

Academic Editor: You Seung Rim

Received: 27 February 2022
Accepted: 16 April 2022
Published: 20 April 2022

1. Introduction

In recent years, the application of flexible electronics in the field of communication has developed rapidly. Driven by the new generation of information and communication network technology, the manufacturing industry will rapidly complete the digitization, networking, and intelligence transformation, of which the key source technology is a perception [1]. Some flexible electronics are required to have a perception function to truly allow perception to be free from multidimensional space. Flexible perception can subversively change the rigid physical form of traditional information devices and systems, realize the efficient integration of information with people, objects, and the environment, realize the flexibility of information acquisition, processing, transmission, display, and even energy, and realize "all things are interconnected" [2]. Although there are many kinds of flexible electronic devices, and their applications are wide, the basic structures of most devices are similar. Generally speaking, flexible electronic devices are mainly composed of flexible substrates, electrodes, functional components, and packaging materials [3]. To achieve flexibility of the whole device, these functional materials need to be flexible enough under the premise of stable operation. Flexible electrodes are the core components of flexible electronics. They need not only good conductivity, but also excellent flexibility. Flexible electrodes usually match the mechanical flexibility of the substrate well. However, when large deformation occurs, the flexible electrodes break, which invalidates flexible electronics and restricts the further development of flexible electronics technology [4]. Therefore, there is a pressing scientific need to study flexible stretchable electrodes, which can guarantee high stability and reliability when the material of the electrodes undergoes large deformations [5,6].

The key to flexible stretchable electrodes is achieving flexibility and increasing their service life under repeated cyclic loading with certain stress (>>1%) [7]. Stretchability refers to the ability of flexible stretchable electrodes to maintain electrical conductivity under mechanical deformation. In the field of materials, it is a research hotspot to prepare materials with more flexibility and even stretchability; in mechanics, geometric graphic design is a shortcut to achieving overall structural stretchability; in micro-nano processing, the development of preparation process matching various new materials is a necessary step to achieve large-scale preparation.

This paper first introduces the concept, performance parameters, and functional adjustment of flexible stretchable electrodes, and then systematically introduces the research results of the flexible stretchable electrode from three aspects of material, technology, and structure. In the first part, the common flexible materials for flexible stretchable electrodes are summarized; In the second part, flexible stretchable electrodes with different stretchable structures are summarized. Electrodes with stretchable structures can reduce their limitations in the preparation of materials; the third part summarizes the current preparation techniques of flexible stretchable electrode materials and classifies them according to different principles in the preparation process. Finally, the existing problems of flexible stretchable electrodes are analyzed, and future development is prospected.

2. Concept, Performance, and Functional Adjustment of Flexible Stretchable Electrode

2.1. Concept

As shown in Figure 1, the ordinary electrode belongs to a rigid physical form and does not have flexibility or extensibility; the structure of the flexible electrode is integrated, which is flexible, but not stretchable; the flexible stretchable electrode is flexible and stretchable. Ordinary electrodes are generally prepared by mixing active materials, adhesives, and conductive agents, grinding them evenly, and then coating metal collectors with them. Due to the heterogeneous interface between the collector and the active material layer, such electrodes are prone to fall off or powder when folded or bent, leading to failure of the electrodes [8]. The development of flexible electronics requires high capacity, high stability, and high flexibility of the electrode materials. Flexible electrodes (Figure 1) are mainly made up of conductive flexible self-supporting electrodes or by loading active materials onto flexible conductive materials [9]. Morphologically, the perception of multidimensional space is characterized by repeated stretching, compression, folding and torsion. Flexible electronics are required to maintain their original functions in any of the above-mentioned shapes, and it is not enough for electrodes to be flexible only.

The flexible electrodes that retain their function despite a strain of 10% are referred to as flexible stretchable electrodes, based on the elastic limit of the human skin [10]. In addition to being highly flexible, conductive, and loaded with active materials, flexible stretchable electrodes also need to meet the requirements of stretchability, strong adhesion, surface modifiability, and biocompatibility [11]. In the future, our perception will be greatly expanded by electronics built into our clothes and accessories, attached to our skin, and even implanted into our bodies, enabling us to blend highly efficiently with the perceived information. In addition, due to advances in flexible materials and flexible electronic products, great progress has been made in bioheuristic flexible robot technology [12–17].

From a biomedical perspective, this biocompatible electronic product, including wearable electronic devices [18,19], epidermal devices [20–24], and implantable electronic devices [25–28] (Figure 1), plays an increasingly important role in human health monitoring, which will significantly change the future of health care and our relationship with electronics; from the perspective of smart robots, a biomimetic soft robot with skin-like sensors and soft actuators (Figure 1) is expected to interact with human beings and their surroundings, significantly enhancing security, sensitivity, and adaptive responses, to achieve a complete human-machine cycle; from the perspective of the future industrial internet, it can achieve the multidimensional and high-fidelity representation of production feature data, and ultimately achieve the accurate construction and free regulation of the Internet through

the establishment of the model database, the classification of production elements, and the correlation between each model and each other.

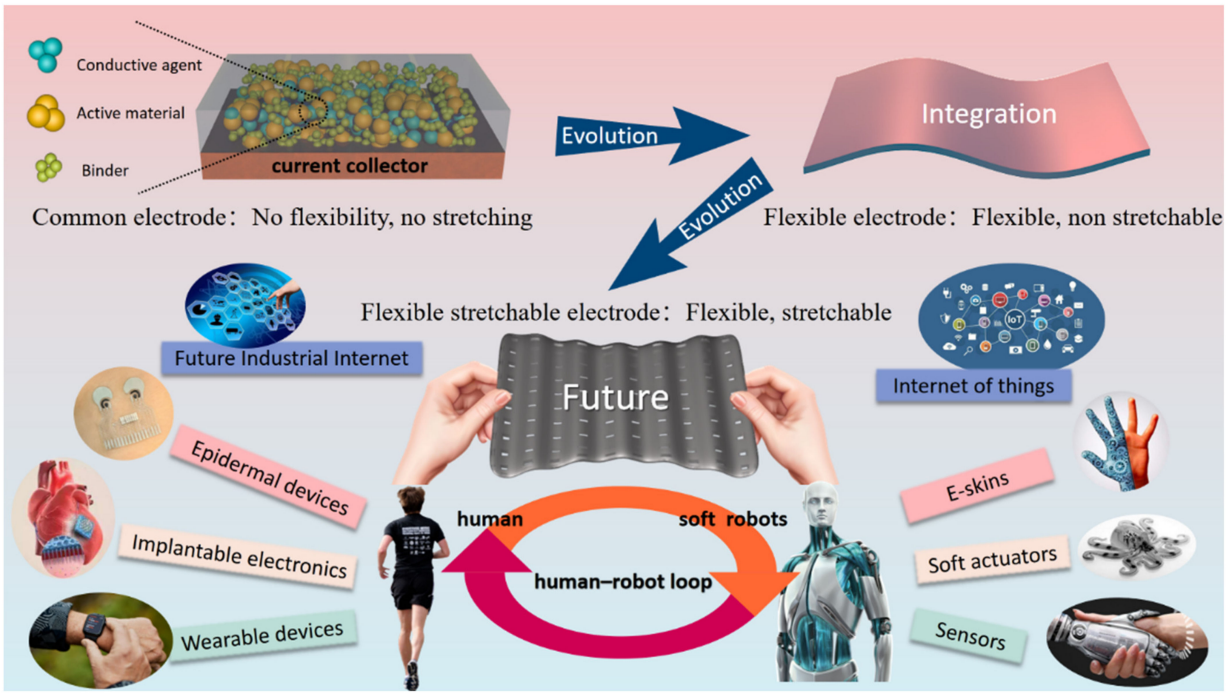

Figure 1. Development and prospects of flexible stretchable electrodes. Image for "Epidermal devices": Reproduced under the terms of the CC-BY Creative Commons Attribution 4.0 International license (https://zhuanlan.zhihu.com/p/351604777 (accessed on 2 April 2022)) [29]. Image for "Implantable electronics": Reproduced under the terms of the CC-BY Creative Commons Attribution 4.0 International license (https://www.sohu.com/a/279876829_100005926 (accessed on 2 April 2022)) [30]. Image for "Wearable devices": Reproduced under the terms of the CC-BY Creative Commons Attribution 4.0 International license (https://www.sohu.com/a/450746414_114822 (accessed on 2 April 2022)) [31]. Image for "E-skins": Reproduced under the terms of the CC-BY Creative Commons Attribution 4.0 International license (https://www.sohu.com/a/226167819_460436 (accessed on 2 April 2022)) [32]. Image for "Soft actuators": Reproduced under the terms of the CC-BY Creative Commons Attribution 4.0 International license (http://www.chinainc.org.cn/show-14-104546-1.html (accessed on 2 April 2022)) [33]. Image for "Sensors": Reproduced under the terms of the CC-BY Creative Commons Attribution 4.0 International license (https://www.sohu.com/a/253004726_100101359 (accessed on 2 April 2022)) [34].

2.2. Performance

Conductivity is one of the key indices of the electrical properties of flexible stretchable electrodes, which is related to the composition and structure of the material itself. The flexible stretchable electrode with good performance should show low resistance under the maximum operating strain, and the resistance value is almost unchanged during the cycle. Generally, the square resistance is the resistance value on the unit thickness and unit area of conductive materials, and the unit is Ω/sq or Ω/□. For a thin square film electrode with side length l and thickness d, the square resistance can be expressed as: $R_S = \frac{\rho l}{ld} = \frac{\rho}{d}$. Where the resistance R_S (Ω/sq) is equal to the volume resistivity ρ (Ω·m) divided by thickness d, conductivity σ (S/m) is the reciprocal of volume resistivity. It can be seen from the above formula that the block resistance is independent of the sample size, directly proportional to the resistivity and inversely proportional to the thickness of the film sample. It is a parameter comprehensively reflecting the conductivity of the film. Among them, "square" has no practical significance, but is used to distinguish it from the resistance between two points.

As shown in Figure 2, the deformation types of flexible electronics in different use scenarios can be divided into bending, twisting, uniaxial tensile deformation, biaxial tensile deformation, circumferential tensile deformation, etc. As an indispensable part of flexible electronics, electrode materials can also withstand external forces and resist deformation, which we call the mechanical properties of materials. Under the action of external forces, the shape and size of the material change, which is called deformation. When a material is deformed, the relative positions and distances between molecules or ions within it change, and additional internal forces between atoms and molecules will be generated to resist external forces, and an attempt is made to restore to the state before the deformation. When the balance is reached, the additional internal forces are equal in size, and opposite in direction, to the external forces. The additional internal force on a material's unit area is called stress, and its value is equal to the external force on a unit area; that is, $\sigma = F/A$, where σ is stress, F is external force, and A is area. In the International System of Units, the unit of stress is N/m^2, which is also written as Pa. If the area before the material is stressed is A_0, then $\sigma_0 = F/A_0$ is the nominal stress. If the material is compressed by force and the area is A, $\sigma_T = F/A$ is the true stress. Nominal stress is commonly used in practice, and there is little difference in numerical values between materials with small deformation.

Figure 2. Deformation types of flexible electronics. Image for "Twisting": Reproduced under the terms of the CC-BY Creative Commons Attribution 4.0 International license (https://m.sohu.com/a/293201739_427506 (accessed on 2 April 2022)) [35]. Image for "Bengding": Reproduced under the terms of the CC-BY Creative Commons Attribution 4.0 International license (https://www.cechoice.com/article/34160_3.html (accessed on 2 April 2022)) [36]. Image for "Radial": Reproduced under the terms of the CC-BY Creative Commons Attribution 4.0 International license (https://www.ctn1986.com/index.php?c=content&a=show&id=75006 (accessed on 2 April 2022)) [37]. Image for "Biaxial": Reproduced under the terms of the CC-BY Creative Commons Attribution 4.0 International license (https://www.sohu.com/a/124592411_119659 (accessed on 2 April 2022)) [38]. Image for "Uniaxial": Reprinted with permission from [39]. Copyright 2018 Nature materials.

Strain is used to characterize the relative displacement of particles within a material under stress. For isotropic materials, there are three basic types of strain: tensile strain ε, shear strain γ and compression strain Δ. For ideal elastic materials, elastic deformation occurs under stress. The relationship between stress and strain obeys Hooke's law, that is, the stress σ is proportional to the strain ε: $\sigma = E \cdot \varepsilon$. The scale factor E in the formula is called the elastic modulus. Elastic modulus is the stress at which a material undergoes unit strain, which indicates the resistance of the material to deformation. The larger the E, the

less likely the material will be to deform, indicating greater stiffness of the material. The unit of elastic modulus is the same as the unit of stress, both N/m^2.

2.3. Functional Adjustment

Flexible electrodes can be divided into three levels according to their mechanical properties. The lowest level is bendable and coileable, the second level is plastic stretchable, and the highest level is flexible stretchable. As far as the degree of preparation difficulty, the higher a material is in the hierarchy, the more difficult the preparation. Concerning application requirements, the higher a material is in the hierarchy, the broader the scope of application. The development of flexible stretchable electrodes is a hotspot and difficult point in current research. A large stretching range (>1%) requires higher performance and structure of materials [40]. Flexible stretchable materials are usually prepared by two methods: one is to develop a new type of flexible stretchable material [41], and the other is to process the material into a geometric structure with scalability. As shown in Table 1, the process complexity of the two methods is different and the flexible stretchable materials prepared have different mechanical properties.

Table 1. Comparison of two different types of flexible stretchable electrodes.

Type	Flexible Form	Patterning	Stretch Direction	Fatigue Resistance	Process Complexity and Cost
Flexible stretchable material	Flexible material	Easily	Multiple Directions	Moderate	Simple, large-area preparation and low cost
Structural design of stretchable materials	Structural deformation	Difficulty	Uniaxially/Biaxially	Good	Complex, small size, high cost, and high equipment requirements

Flexible stretchable electronic materials can be divided into two categories: flexible stretchable materials and flexible stretchable composite materials. Flexible stretchable materials refer to materials that have certain stretchability when conductive polymers (Figure 3a), metal nanostructures (Figure 3b), graphene (Figure 3c), and carbon nanotubes (Figure 3d) are processed into thin films, fibers, and blocks. Flexible stretchable composite materials refer to the preparation of stretchable composite materials by loading the active materials which are not stretchable on the flexible stretchable materials. Electrodes made of flexible stretchable electronic materials have excellent stretchability and tensile restorability, but their shortcomings are limited by the material itself, and their conductivity and energy density are low.

It is difficult for flexible stretchable materials to maintain absolute stability of resistance during stretching, and it is unavoidable that conductive materials undergo micro-structure fracture when they recover from stretching. Due to the low electrical stability of flexible stretchable materials, scientists have proposed geometric structures to achieve the stretchability of materials [42,43]. Conductors are fabricated into scalable geometries such as mesh [44] (Figure 3e), fabric [45] (Figure 3f), wavy [46] (Figure 3g), snake [47] (Figure 3h), and spiral [48] (Figure 3i) to form flexible stretchable electrodes. When an electrode with a stretchable structure is stretched, the conductor material can cleverly use its geometric shape to buffer the external stress, thus maintaining a stable conductivity at a larger stretch length and even lower than the resolution limit.

After the requirement of flexible stretchability was achieved in mechanical performance, with further research on flexible stretchable electrodes it was applied to epidermal electronic devices and implantable electronic devices [49,50], which puts forward new requirements for flexible stretchable electrodes. To achieve a close skin fit, flexible stretchable materials need to have a lower Young's modulus (<260 kPa [51]) than the human epidermis. In addition, long-time wear puts higher requirements on the biocompatibility and wearing comfort of the materials.

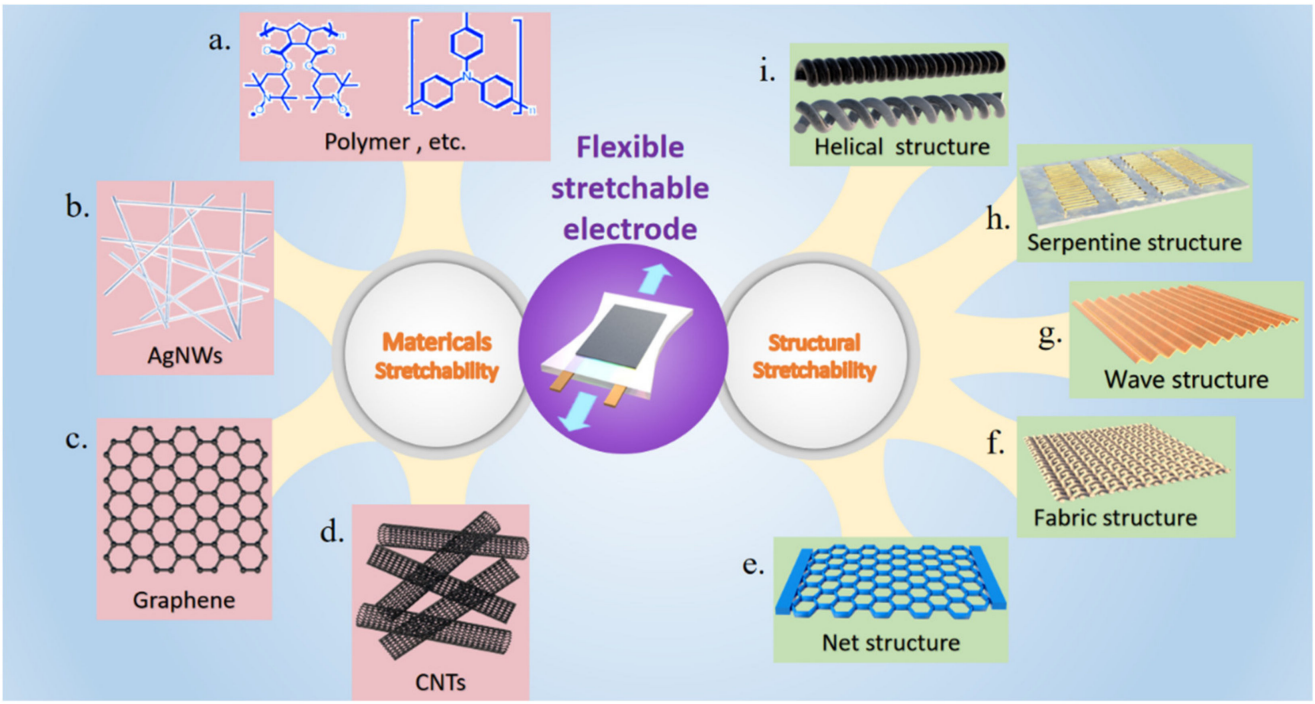

Figure 3. Flexible stretchable materials: (**a**) conductive polymers, (**b**) metal nanostructures, (**c**) graphene and (**d**) carbon nanotubes, and stretchable structures: (**e**) mesh, (**f**) fabric, (**g**) wavy, (**h**) snake, and (**i**) spiral.

3. Varieties of Flexible Stretchable Electrode Substrate Materials

Flexible stretchable electrode materials usually need to be combined with active and base materials to improve their mechanical and electrical properties. As shown in Table 2, the conductivity of flexible stretchable materials is compared. Some active materials can act as collectors to make the polymer conductive and have electrochemical activity to contribute capacitance. Some active materials can only be plastic deformed, but not resilient. Loading this material on an elastic substrate yields a composite material which has the conductivity, electrochemical activity, and elasticity of the active material. From the base material, flexible stretchable electrode materials include carbon-based electrode materials, elastic polymer-based electrode materials, paper-based, fabric-based, and metal-based. The flexible stretchable electrodes made of different substrate materials are listed in detail below.

Table 2. Conductivity and tensile rate of flexible electrode materials.

Material	Conductivity	Ultimate Tensile Rate	Reference
Solution-styrene butadiene rubber/Carbon black particles	40 S/m	200%	[52]
Graphene foam/PDMS	72 S/m	60%	[53]
SBR/rGO-CNT	3.62 S/cm	5%	[54]
Fluorinated rubber/SWNTs	100 S/m	100%	[55]
PUS-AgNW-PDMS	19.2 S/cm	100%	[56]
Single covered yarn-AgNW fiber	4018 S/cm	500%	[57]

3.1. Carbon-Based Electrode Materials

From Table 3, carbon-based materials are widely used in the preparation of flexible stretchable electrodes due to their high specific surface area, high conductivity, and good chemical stability. In particular, carbon nanotubes (CNTs) and graphene with unique structures are well suited for flexible stretchable devices [58]. One-dimensional CNTs can be directly made into full-carbon thin films or fiber electrodes, and graphene can also be

used to synthesize graphene thin films, graphene fibers, etc [59–63]. In addition, nanocarbon fibers, carbon paper and the composite of two carbon materials have been applied to stretchable electrode materials by researchers because of their high specific capacitance and good cyclic stability.

Table 3. Study on flexible stretchable electrodes made of carbon-based materials.

Electrode Material	Preparation Method	Ultimate Tensile Rate	Wear-and-TearLife	Reference
CNT array	Chemical vapor deposition	30%	Maintain performance after 30% tensile strain	[64]
CNTs fibers	Pre-stretching	100%	Maintain performance after 100% cyclic tensile strain	[65]
CNTs nano-tablets	Chemical vapor deposition	100%	Maintain 84% performance after 200 tensile cycles at 100% tensile strain	[66]
Graphene paper	Solution method	800%	Maintain performance after 1000 tensile cycles	[67]
Graphene/CNTs fibers	Wet-spinning	800%	Maintain 77% performance at 800% tensile strain	[68]
MnO_2 nanowires/Cellulose nanofibers/CNTs	Hydrothermal method	500%	Maintain 96% performance at 400% tensile strain	[69]
MnO_2/Graphene/Ni	Chemical vapor deposition	100%	Maintain 92% performance at 100% tensile strain	[70]
MoS_2/CNTs	Chemical vapor deposition, Immersion method	240%	Maintain performance after 240% tensile strain	[71]
CNTs/Silver nanoparticles	Electrospinning	550%	Maintain performance at 550% strain tensile range	[72]

Xiao et al. used the synergy between carbon nanotubes with good mechanical properties and vanadium nitride (VN) with the mesoporous structure to build a flexible membrane electrode with a self-supporting structure [73]. Niu et al. prepared a flexible composite film electrode with a "skeleton/skin" structure [74]. The electrode takes a single-wall carbon nanotube (SWCNT) with a network structure as the "skeleton", and polyaniline (PANI) is grown on the skeleton as the skin layer through in-situ chemical polymerization of aniline (ANI) monomer (Figure 4a). Xu et al. prepared a composite material with high conductivity and high current capacity based on RGG (reduced giant graphene) doped with silver ions or silver fibers (Figure 4b), which can also be used to prepare flexible stretchable electrodes [75]. Zhou et al. designed a folded carbon electrode using pencil and sandpaper, which showed good tensile properties and was successfully applied to wearable electronic devices [76]. For the current electrode materials, the stretchability mostly comes from carbon materials, and most carbon materials have certain advantages and disadvantages. People will continue to develop new carbon or explore simple and economical new technologies and processes.

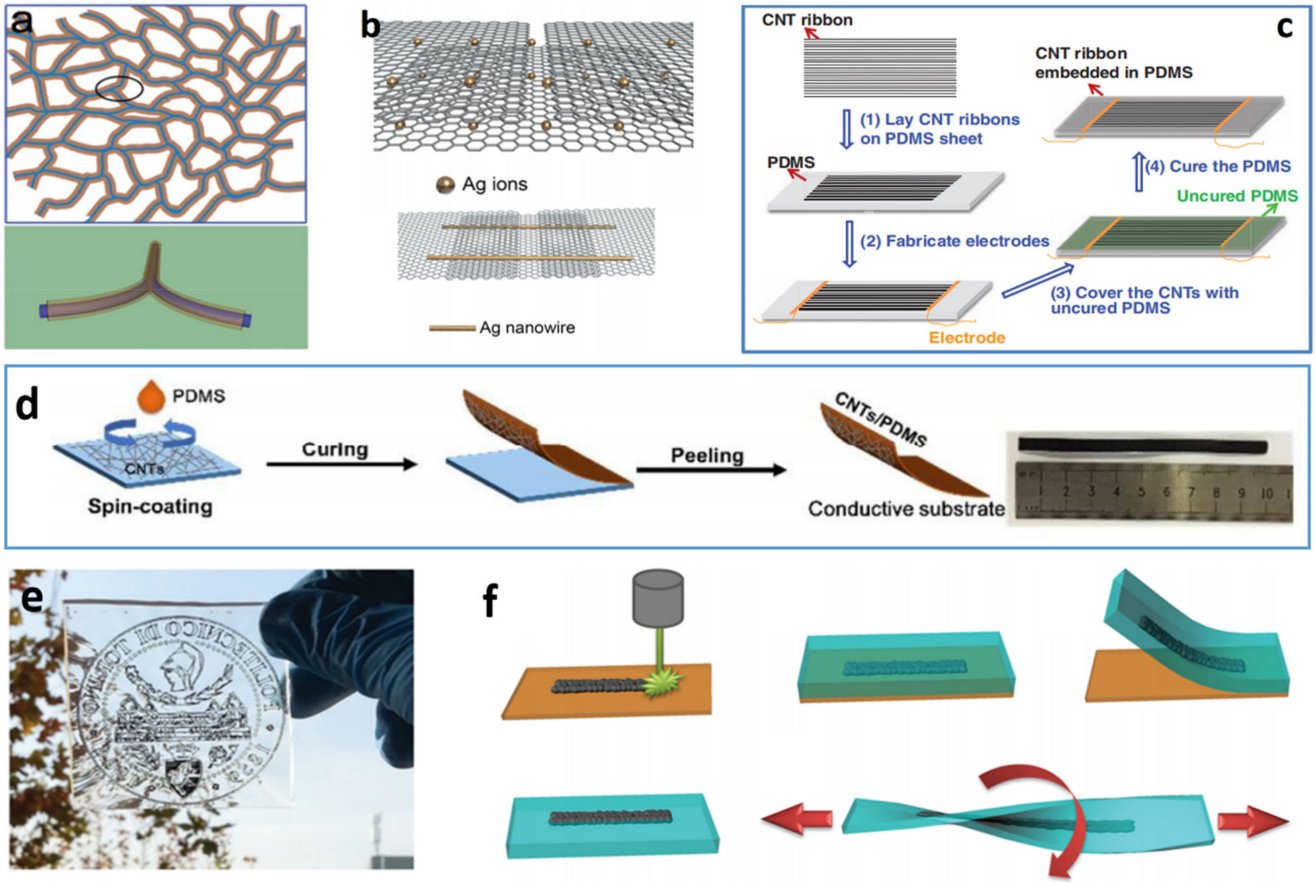

Figure 4. The sketch diagrams of a single layer film based on (**a**) continuous SWCNT/PANI reticulate structure. Reprinted with permission from [74]. Copyright 2012 Energy & Environmental Science. (**b**) Schematic images of the molecular doping mechanism by Ag ions in HI-reduced RGG-Ag fibers (up) and complex mechanism by AgNWs in VC (Vitamin C)-reduced RGG-Ag fibers (down). Reprinted with permission from [75]. Copyright 2013 Advanced Materials. (**c**) Schematic illustration of the process to embed CNT ribbons in PDMS. Before encapsulating the CNTs, electrodes connected to the two ends of CNT ribbons are fabricated to enable conductivity measurement. Reprinted with permission from [77]. Copyright 2010 Advanced Materials. (**d**) Schematic illustration for the fabrication process of stretchable CNTs/PDMS substrates. Reprinted with permission from [78]. Copyright 2018, Nanoscale. (**e**) PDMS and LIG/PDMS substrate. Reprinted with permission from [79]. Copyright 2016 Advanced Energy Materials. (**f**) 3D scheme of the LIG transfer onto PDMS substrate and its elastomeric properties. Reprinted with permission from [80]. Copyright 2016 Advanced Energy Materials.

3.2. Polymer-Based Electrode Material

Table 4 summarizes commonly used flexible stretchable electrode materials based on elastic polymers, including polydimethylsiloxane (PDMS), polyurethane (PU), thermoplastic copolymer (Ecoflex), and PEDOT:PSS. Polymer materials can be made into thin films by solution coating, showing good elasticity and tensile recovery, and can be stretched and used repeatedly. It can also be seen from Table 4 that there are many materials that can convert polymers into good fluid collectors and be used in various flexible electronics. The polymer is evenly coated on the carbon nano materials, which can be peeled off after curing into a film; metal materials, such as gold, silver, and copper, can be made into metal nanoparticles or metal nanowires, and then doped/deposited with polymers to prepare highly conductive current collectors; conductive polymers (PPy, PANI, polythiophene) can directly deposit a conductive layer on the polymer surface by electrodeposition.

Table 4. Application of flexible stretchable electrode based on polymer in electronics.

Electrode Material	Preparation Method	Ultimate Tensile Rate	Wear-and-Tear Life	Application	Reference
V_2O_5/PEDOT/PDMS	Spin coating	50%	Maintain 85% capacitance after 100 tensile cycles	Supercapacitor	[81]
SWCNTs/BNNT/PDMS	Dry pressure	50%	After 1000 tensile cycles at 50% strain, the capacitance increases by 25%	Supercapacitor	[82]
Graphene/PDMS	Chemical vapor deposition	1.3%	Maintain performance after 300 cycles	Pressure sensor	[83]
AgNWs/PDMS	Coating	70%	70% tensile rate strain	Strain sensor	[84]
PPy/PU	Chemical polymerization	100%	Maintain 90% capacitance after 1000 times of 100% tensile strain	Supercapacitor	[85]
SWCNTs/Ecoflex	Coating	60%	After 1000 charge discharge cycles, the capacitance remains 97.4%	Supercapacitor	[86]
Polyaniline-multi-carbon nanotube/PEDOT:PSS	Electropolymerization	50%	The CV Curve remained unchanged after 30 tensile cycles at 50% strain	Supercapacitor	[87]
SWCNTs/PEDOT:PSS	Spin coating	100%	Maintain performance after 1000 times of 20% tensile strain	Strain sensor	[88]
ZnS:M^{2+}(Mn/Cu)@ Al_2O_3/PDMS	Print	20%	Maintain performance during 10,000 bending and stretching cycles	Touch sensor	[89]
TPU/AgNWs/rGO	Spray	200%	Maintain performance after 200% tensile strain	Nano Generator	[90]
PDMS/CNTs fibers/Graphene nanoplates	Spin coating	50%	Maintain performance after 1000 stretch cycles	Strain sensor	[91]

For example, the conductive filler carbon nanotubes (CNTs) are evenly spread on the glass substrate and then spin-coated with PDMS solution [92]. After cooling and curing, CNTs/PDMS film is peeled off (Figure 4d) to obtain a conductive collector [78]. As shown in Figure 4c, the CNT ribbons can also be directly placed on the surface of PDMS film, and then the electrodes at both ends of the CNT ribbons for conductivity measurement can be prepared. A layer of uncured PDMS can be cast on the CNT/PDMS to make a stretchable CNT/PDMS electrode film [77]. It is worth noting that the interaction between the carbon nanotubes and the PDMS is very strong. Once carbon nanotubes contact the surface of PDMS film, they cannot move by mechanical scratching. Therefore, this kind of electrode solves the problem of poor adsorption between the conductive components and the substrate materials. Laser-induced graphene (LIG) is also an effective conductive material [79]. The stretchable electrode can also be prepared by transferring LIG to PDMS (Figure 4e). As shown in Figure 4f, the manufacturing process of LIG/PDMS electrode is described: a porous LIG pattern was obtained by directly writing a polyimide sheet with a laser; Then PDMS was completely injected into the 3D network, and LIG/PDMS thin film electrode was obtained after curing [80].

Choi et al. embedded transparent retractable silver nanowires/reduced graphene oxide (AgNWs/rGO) electrode wires into polyurethane (PU) dielectric layer on PDMS substrate (Figure 5a), and directly manufactured thin-film electrodes on elastomer substrate to prevent capacitance change caused by stretching [93]. At the same time, PU material is also a polymer with excellent deformation ability, which can be used as an elastic matrix alone. NCNT/PU film was prepared by coating PU onto an N-doped CNT array (Figure 5b) [94]. In addition, researchers have uniformly grown ultra-thin manganese dioxide nanosheets on PU-NCNTs substrate by a simple microwave radiation method and prepared a new electrode with excellent performance [95]. Alternatively, the AgNWs conductive filler can be directly added to the PU elastic matrix to make a flexible AgNWs electrode [96].

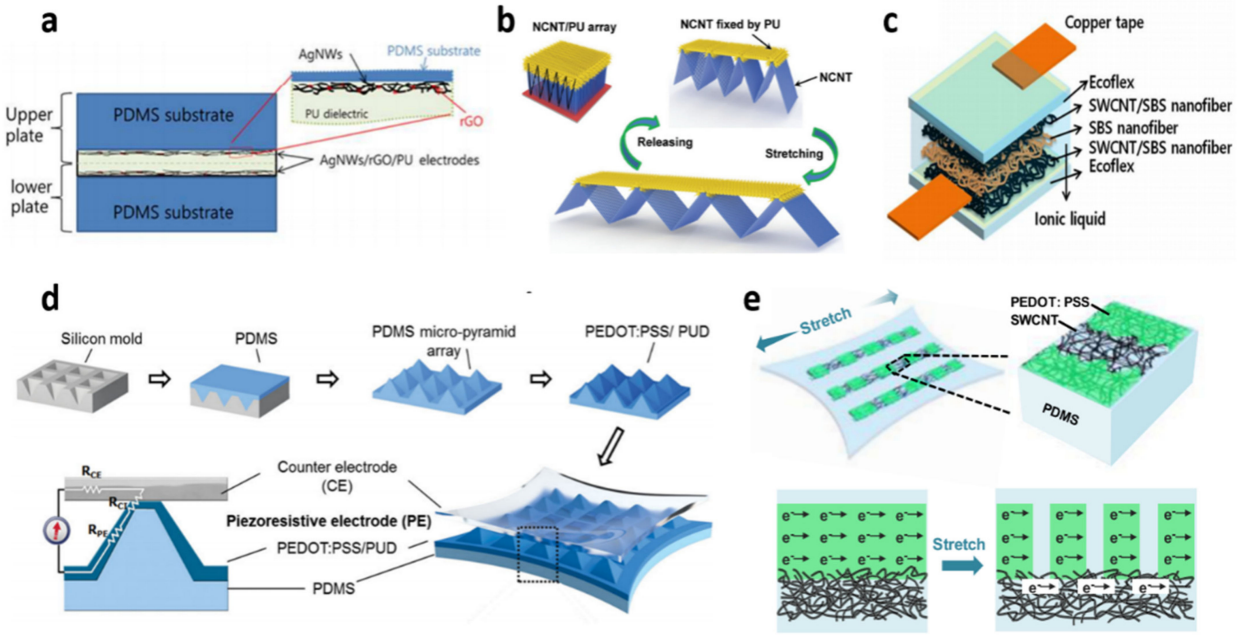

Figure 5. Schematic diagram of PDMS, PU, Ecoflex, PEDOT:PSS substrate flexible electrode. (**a**) Schematic of the transparent and stretchable capacitive sensor. Reprinted with permission from [93]. Copyright 2017 ACS Applied Materials& Interfaces. (**b**) Schematic illustration of the stretching mechanism of NCNT/PU film. Reprinted with permission from [94]. Copyright 2016 Advanced Energy Materials. (**c**) Assembled stretchable supercapacitor device. Reprinted with permission from [97]. Copyright 2018 Nanomaterials. (**d**) A PEDOT:PSS/PUD thin film coating the pyramid surface and circuit model used to derive the sensing principle of the sensor. Reprinted with permission from [98]. Copyright 2014 Advanced Materials. (**e**) Schematic diagram of PEDOT:PSS/SWCNT electrodes and corresponding images showing good stretchability when strain is applied on the electrode. Reprinted with permission from [99]. Copyright 2020 ACS Applied Materials & Interfaces.

PEDOT:PSS, as a conductive polymer, is also frequently prepared into thin films and gels, whose electrical properties are affected by different morphologies [100]. Choong et al. copied the micro pyramid PDMS substrate with silicon mold, and grafted PEDOT: PSS/PUD composite polymer (Figure 5d), and formed the structure of the piezoresistive electrode [98]. Zhu et al. compounded CNT and Ag-doped PEDOT: PSS materials to prepare flexible electrodes with a maximum tensile rate of 480% [101]. Zhao et al. [99] used PEDOT: PSS/SWCNT composite as an electrode and the elastic polymer PDMS was rotationally coated on the PEDOT: PSS/SWCNT electrode array to embed the electrode array in PDMS to obtain a flexible stretchable electrode (Figure 5e). PEDOT:PSS solution can also be spin-coated on SEBS elastomer substrate, and SEBS is selected as the substrate because. Unlike PDMS, its physical properties such as hardness, tensile strength, and elongation are not easy to change [102]. Some researchers fabricated stretchable electrodes such as carbon nanotube [103], multi-walled carbon nanotube (MWCNTs) [104], nanofiber composite (single-walled carbon nanotube/styrene (SWCNT/SBS)) (Figure 5c) [97] on an Ecoflex elastic substrate with an ultimate tensile rate of 60%.

3.3. Electrode Materials with Other Substrates

In addition to the flexible electrodes in the above two substrates, some researchers have used paper-based, fabric-based, metal-based [105–107], etc., to prepare flexible electrodes and electronic devices. For example, a paper-based flexible stretchable electrode was prepared by interfacial polymerization of pyrrole monomers to make its active material, polypyrrole (PPy), grow on the flexible dustless paper [108], or to make CNTs/MnO_2 composite grow on the dustless paper [109]. Hu et al. prepared a highly conductive carbon nanotube/cotton fabric flexible electrode by repeatedly soaking cotton fabric in carbon

nanotube solution by a simple dipping and drying method [82]. Supercapacitors (Figure 6a) made from these conductive textiles show high areal capacitance, up to 0.48 F/cm^2, and high specific energy.

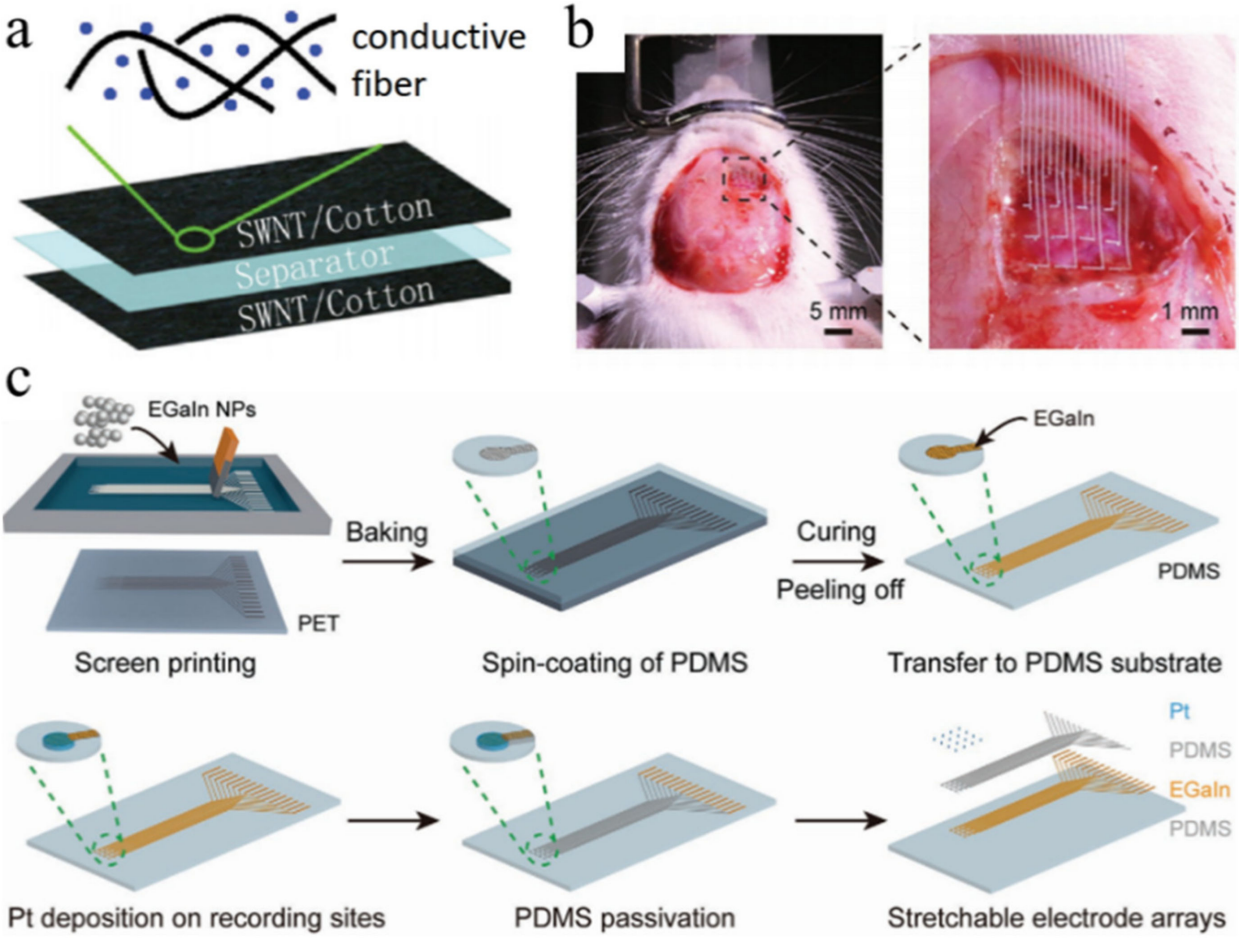

Figure 6. (**a**) SC structure with porous textile conductors as electrodes and current collectors. Reprinted with permission from [82]. Copyright 2010 Nano letters. (**b**) Intraoperative image of the liquid metal neural electrode arrays (left). Intraoperative microphotograph of the 16-channel liquid metal electrodes on the rat brain (right). Reprinted with permission from [110]. Copyright 2021 Small. (**c**) Schematic illustration of the fabrication of SEA using screen printing. Reprinted with permission from [110]. Copyright 2021 Small.

Metal-based electrode materials can be divided into two types, one of which is ordinary metal materials. Xia et al. deposited cobalt hydroxide (Co $(OH)_2$) nanosheets on porous nickel membranes (Ni) by electrochemical deposition [111], using the porosity and flexibility of the nickel membranes to achieve the flexibility of the electrodes. Li et al. directly used gold wire as a flexible matrix to deposit carbon-based materials on the surface of their wire by electrochemical deposition and apply them to flexible supercapacitors [112]. The other is liquid metal. Liquid metals possess high stretchability, good biocompatibility, and excellent electrical conductivity, making them promising candidates for flexible and stretchable electrodes. Liquid metal-based flexible and stretchable electrodes have been used in electronic devices such as nanogenerators [113], diodes [114], and actuators [115]. Ma et al. report a stretchable conductor that is fabricated by simply coating or printing liquid metal onto an electrospun elastomeric fiber mat [116]. The liquid-metal fiber mat shows good biocompatibility and smart adaptiveness to omnidirectional stretching over 1800% strain. Dong et al. proposed that liquid metal electrode has high biocompatibility and good performance in neurite growth and long-term implantation [110]. Figure 6c

demonstrates the fabrication process of stretchable electrode arrays (SEA). As shown in Figure 6b, Due to the high stretchability and softness of the liquid metal electrodes, it could expand and form an intimate interface with the targeted cortical surface.

4. Flexible Stretchable Structure

As shown in Table 5, stretchable structures can be roughly divided into five types: spiral structure, wave structure, fabric structure, mesh structure, and snake structure. Among them, the devices made of spiral structure are generally fibrous and have certain rigidity, such as CNT yarn and stainless steel wire, with a tensile rate ranging from 20% to 400%; the tensile ratio of electrodes or devices with wave structure is generally low, ranging from 30% to 100%; fabric structure can be stretched through special textile structure, such as knitted fabrics and sweaters, with a stretching rate of 20–120%; the ultimate elongation of the electrodes/devices with mesh structure varies greatly from 10% to 2000%, 10% is a normal sponge structure, and 2000% is a paper-cut work of art. The snake-like structure has poor mechanical performance and cannot be restored. The device can only reach 20–30% stretch ratio.

Spiral structure and fabric structure are moderately stretchable, and one-dimensional fiber-like devices can be well integrated into fabric clothing, and the fabric structure itself is a part of fabric clothing, which is an ideal structure form in the development of smart flexible devices [117]. Yu et al. coated carbon nanotube (CNT)@MnO_2 core fibers with gel electrolyte and CNT@PPy one-dimensional fibrous supercapacitor (FSS) with asymmetric nuclear sheaths was prepared by coating the composite film [118], and then an extensible spiral structure was formed by over-twisting (Figure 7a). Chu et al. synthesized layered carbon tubular nanostructures (hCTNs) on the surface of stainless-steel spring (SSS) with helical structure [119], and then polymerized the nano spherical polyaniline (PANI) onto the hCTNs network in turn to form a hCTNs/PANI hybrid stretchable electrode (Figure 7b).

Table 5. Study on flexible stretchable electrodes prepared with different stretchable structures.

Structure	Material	Wear-and-Tear Life	Ultimate Tensile Rate	Reference
Spiral	CNTs yarn/MnO_2/PPy	Maintain 88% capacitance after 200 cycles of stretching under 20% strain	20%	[120]
Spiral	Stainless steel wire/MnO_2/rGO	Maintain 95% capacitance after 3000 cycles under 400% strain	400%	[121]
Spiral	CNTs fibers	Maintain 94% capacitance after 300 stretching cycles	300%	[122]
Wave	CNTs/MnO_2/PPy	Maintain 96% capacitance after 500 stretching cycles	100%	[123]
Wave	Nickel foam/Polyaniline/Graphene	Maintain 95% capacitance after 100 tensile cycles at 30% strain	30%	[124]
Fabric	Ag/PPy/MnO_2	Maintain 86.2% capacitance at 40% strain	40%	[125]
Fabric	CNTs/PPy/MnO_2	Maintain 98.5% capacitance at 21% strain	21%	[126]
Fabric	SWNTs	Capacitance remains unchanged after stretching	120%	[127]
Serpentine	SWNTs	Maintain capacitance after 10 tensile cycles at 30% strain	30%	[128]
Reticular	SWNTs film	Maintain capacitance at 150% strain	150%	[129]
Reticular	PPy/BP/CNTs	Maintain 95% capacitance after 10,000 tensile cycles at 2000% strain	2000%	[130]
Reticular	PPy/CNTs	101% dynamic capacitance after 5000 tensile cycles at 5% strain	10%	[131]

There are two preparation methods for stretchable devices of fabric structure: one is simple, and the stretchable electronic device is prepared by depositing active material on the fabric structure substrate by dipping coating or screen printing. The fabrication process of composite fabric electrodes is shown in Figure 7c and the structural diagram [132] is

applied to supercapacitors. Another is to fabricate fiber-like electrodes and then weave them into fabric structures to obtain stretchable devices, as shown in Figure 7d is a full-solid symmetric yarn supercapacitor with two PEDOT:PSS/CNF (carbon nanofiber)/CF (carbon fiber) electrodes in parallel, and a schematic diagram [133] of its mechanical deformation.

The electrode with wave structure has a simple preparation method and low cost. However, the surface of the prepared material is wrinkled and undulating, so the wearing comfort is poor. There are usually two ways to prepare wavy structures: First, the elastic base is pre-stretched, on which a 2D planar electrode material is deposited, and then the pre-stretched elastic base is released [134] (Figure 8a). The electrodes deposited on it show an undulating wavy structure. The electrodes deposited on it will show an undulating wavy structure. Based on the above method, the Xie group [135] found that porous graphene can be wavy by pre-stretching the substrate, thereby improving the defect of porous graphene with poor stretchability. The wavy PANI/Graphene electrodes were then prepared and packaged to obtain a stretchable supercapacitor.

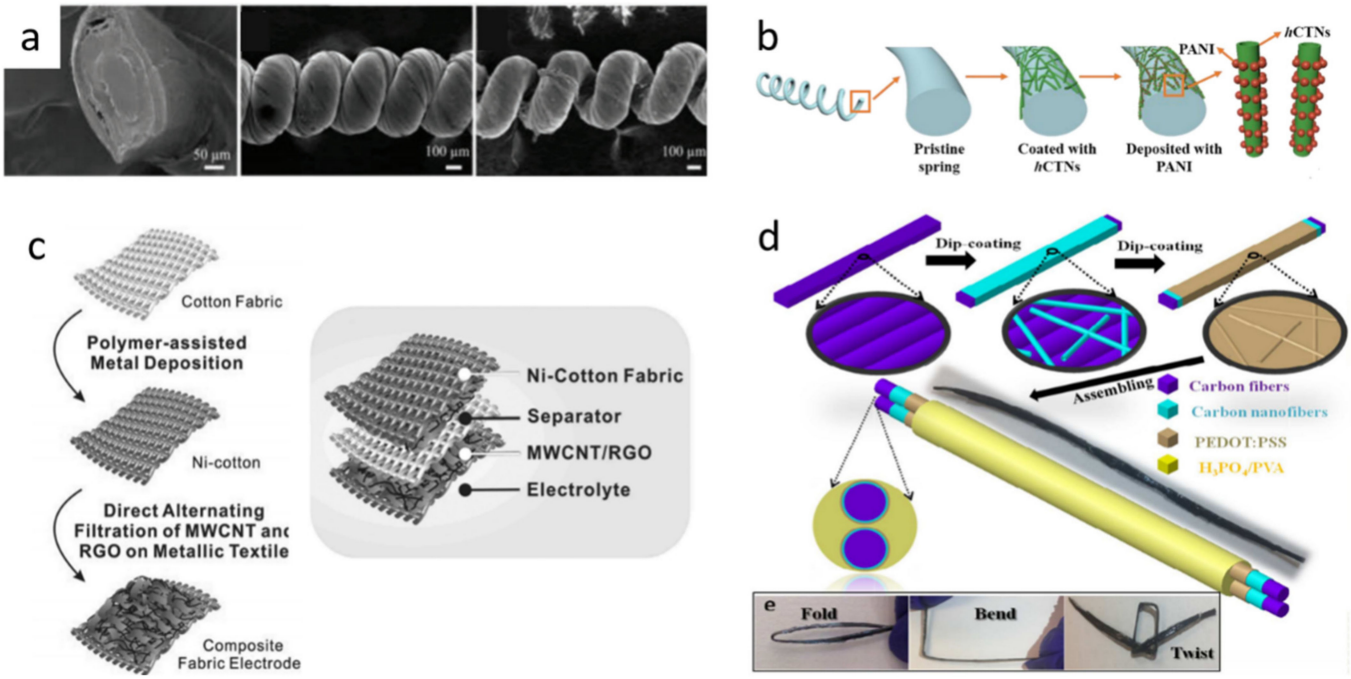

Figure 7. (**a**) SEM image for a cross-section of the fiber supercapacitor and the stretchable FSS at 0% and 50% strain. Reprinted with permission from [118]. Copyright 2016 Advanced Energy Materials. (**b**) A schematic presentation of synthesizing hCTNs/PANI composites onto the surface of SSS substrates. Reprinted with permission from [119]. Copyright 2018 Chemical Engineering Journal. (**c**) Schematic diagram of the Ni-coated cotton fabrics for preparing composite fabric electrodes and structure diagram of high-performance supercapacitor. Reprinted with permission from [132]. Copyright 2017 Advanced Materials. (**d**) Fabrication process of the all-solid-state symmetric yarn supercapacitor composed of two PEDOT:PSS/CNF/CF electrodes in parallel, and (**e**) a photograph of the yarn supercapacitor under various mechanical deformations, such as folding, bending, and twisting. Reprinted with permission from [133]. Copyright 2017 ACS Nano.

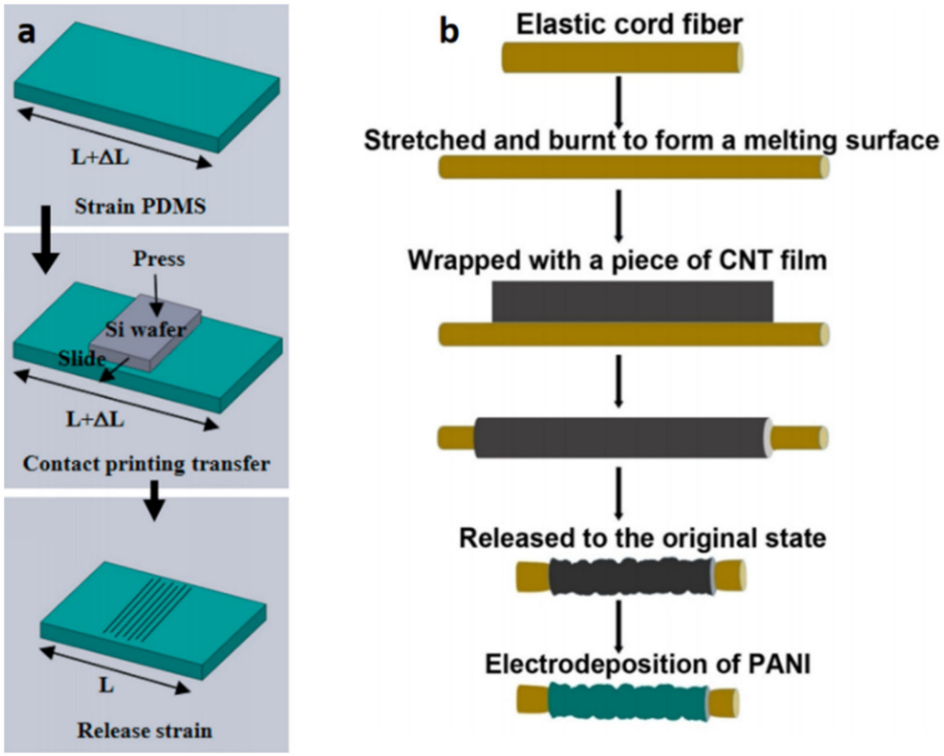

Figure 8. (**a**) Schematic of the strain−release assembly method. Reprinted with permission from [134]. Copyright 2012 ACS Applied Materials & Interfaces. (**b**) Schematic illustration of the fabrication process of the stretchable fiber-like electrode. Reprinted with permission from [136]. Copyright 2018 Chemistry Select.

Secondly, there are fiber wavy electrode materials. Pre-stretch the elastic yarn and wrap the electrode material around it. After the elastic yarn is released, the wavy electrode material is obtained. For example, Ren et al. proposed a method to fabricate composite electrodes by tightly combining CNT/PANI composite membranes with elastic filaments [136]. The wavy structure can be obtained by stretching the elastic filaments, attaching them to the CNT membranes, and then releasing them (Figure 8b). Another stretchable supercapacitor with a pre-stretching strategy introduced PEDOT into SWCNT thin film to obtain (SWCNT/PEDOT) hybrid fiber [137]. A biaxially stretchable supercapacitor with a unique structure is obtained by combining hybrid fibers with PDMS and obtaining wavy fiber electrodes under the pre-elongation strategy.

The mesh structure and the S-shaped structure have the best stretchability and can reach thousands of elongation rates, but they require the high maneuverability of materials [138,139]. If the stretchable structure has no resilience after stretching, it is difficult to restore to its original state, which limits its practical application. Kim et al. produced a supercapacitor consisting of two-dimensional planar SWCNT electrodes and ionic gel electrolytes [140], which rely on narrow, long snake-shaped metal interconnects to ensure mechanical stability during deformation. The micro-supercapacitor array shows good performance when stretched up to 30% (Figure 9a). Yun et al. [141] designed a dual-axis micro-capacitor (MSC) array with integrated solar cells (SCs) and strain sensors (SS), as well as a snake-like interconnect structure and a stretchable substrate (Figure 9b). Two-dimensional scalable electronic devices have limited specific surface area capacitance and are incompatible with three-dimensional wearable devices. To overcome this limitation, it is urgent to develop three-dimensional stretchable electronic devices with higher quality loads and customizable shapes.

The Pu team [142] found that a scalable micro supercapacitor (MSC) on a 3D honeycomb-like PDMS substrate can maintain its capacitance stability even when subjected to large

strains such as stretching, bending, and warping. The stretchability of the MSC array system is also controlled by the distortion of the honeycomb-like PDMS structure (Figure 9c). Inspired by the honeycomb lamp structure, the Lv team [143] proposed a three-dimensional scalable supercapacitor with an expandable honeycomb structure, whose electrodes were prepared by electrodeposition of porous PPy/black phosphorus oxide (BPO) composite onto carbon nanotube (CNT) thin films. Figure 9d shows the manufacturing process of the capacitor. Based on the manganese dioxide nanowire composite electrodes, the team also developed an editable supercapacitor with customizable shape and stretchability [144] (Figure 9e). Its shape is more complex and finer, and therefore it is more controllable.

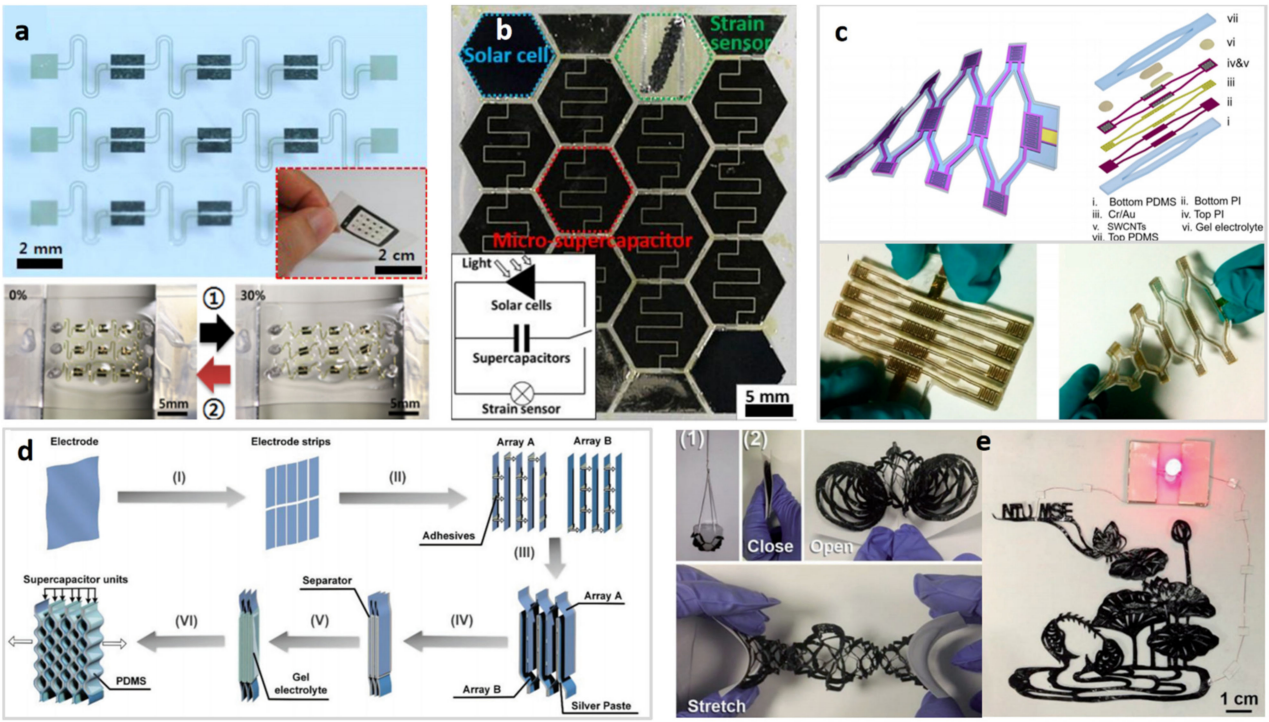

Figure 9. (**a**) Photograph of a stretchable 2D planar micro-supercapacitors array on a PDMS substrate without electrolyte (Inset image shows the tape-transferred micro-supercapacitor array) and photographs of the entire device of the 3 × 3 micro-supercapacitor array before (Lower left) and after application of strain of 30% (Lower right). Reprinted with permission from [140]. Copyright 2013 ACS Nano. (**b**) Optical image of the biaxially stretchable MSC array with integrated SS and SCs. The inset shows the circuit diagram. Reprinted with permission from [141]. Copyright 2018 Nano Energy. (**c**) Schematic illustration of a stretched and bent device; exploded view of various structural layers of the device; digital photograph of a compressed (Lower left) and twisted and stretched (Lower right) device. Reprinted with permission from [142]. Copyright 2016 ACS Nano. (**d**) Schematic drawing of the fabrication process for 3D stretchable supercapacitors: (I) The PPy-based electrodes was first to shaped into identical electrode strips with predefined shapes; (II) the electrode strips were adhered by adhesives (PVA/H_3PO_4 gel electrolyte) and connected into two electrode arrays (array A and array B); (III) the edge of the array A and B were coated with a silver paste to ensure conductive connections, and then the array A were intersected with array B; (IV) two arrays were separated by a thin nanocellulose separator (about 20 µm). (V) Array A and B were coated with PVA/H_3PO_4 gel electrolyte to form a 3D stretchable supercapacitor with expandable honeycomb structures. (VI) The 3D stretchable supercapacitor was stretched to form a honeycomb structure and packaged with a thin-layer PDMS. Reprinted with permission from d. Copyright 2018 Advanced Materials. (**e**) Supercapacitor basket ((1), supercapacitor pyramid conformal to 3D objects), four pyramid-like supercapacitors (2) connected in series, forming a stretchable pop-up spring structure. (Right) Four symmetric supercapacitors connected in series and tailored into more complicated and delicate paper-cutting. Reprinted with permission from [144]. Copyright 2017 Advanced Materials.

5. Preparation Process

Selecting an appropriate method to prepare flexible and conductive electrodes is crucial for the construction of flexible electronics. The preparation methods reported in the literature are generally divided into the following categories, and the advantages and disadvantages are summarized in Table 6.

Table 6. Advantages and disadvantages of different preparation methods.

Preparation Process	Advantages	Disadvantages
Phase conversion method	Simple operation, efficient and mean pore diameter	Small scope of application
Chemical oxidation polymerization method	Simple operation, fast synthesis speed and large output	The control of polymerization process is difficult
Electrochemical deposition	Plasticity, uniformity, controllable thickness and uniform surface	High cost
Chemical vapor deposition	Good wrapping plating, Controllable thickness	High by-products, Not bendable
Physical vapor deposition	Wide application range, Environmental protection	Poor uniformity
Electrospinning technology	Easy to operate, one step shaping, good uniformity and mass production	Low productivity, High cost, Relative product instability
Coating, Printing, and Spraying	Simple equipment, easy process control and easy doping	Inefficient, Poor uniformity
3D printing	Ability to create any geometric shape, Capability of highly accurate conformal deposition, Good mechanical bending	High cost
Vacuum suction	Controllable composition, controllable thickness and well-distributed	Weak mechanical strength

5.1. Phase Inversion Methods

Phase inversion methods can be divided into precipitation from vapor phase, precipitation by controlled evaporation, thermally induced phase separation, and immersion precipitation. One or more conductive fillers are mixed with dispersants and flexible base materials to form conductive nano-composite materials, which are then prepared into a conductive film by scratching or rotating coating. This method is one of the most common methods to prepare flexible electrodes, which is easy to operate and has the advantage of high yield. Zhao et al. [145] facilitated the preparation of polyethersulfone/activated carbon (PES/AC) flexible membrane electrodes by the phase inversion method. The transparent and uniform solution was dispersed into the membrane by rotating coating technology and immersed in deionized water immediately in a condensation bath, thus enabling good contact between the component materials (Figure 10a). Sachse et al. [146] prepared a transparent conductive nano-silver wire electrode with 85% visible light transmission and 15 $\Omega \cdot \square^{-1}$ average square resistance on glass by immersion coating, as shown in Figure 10b.

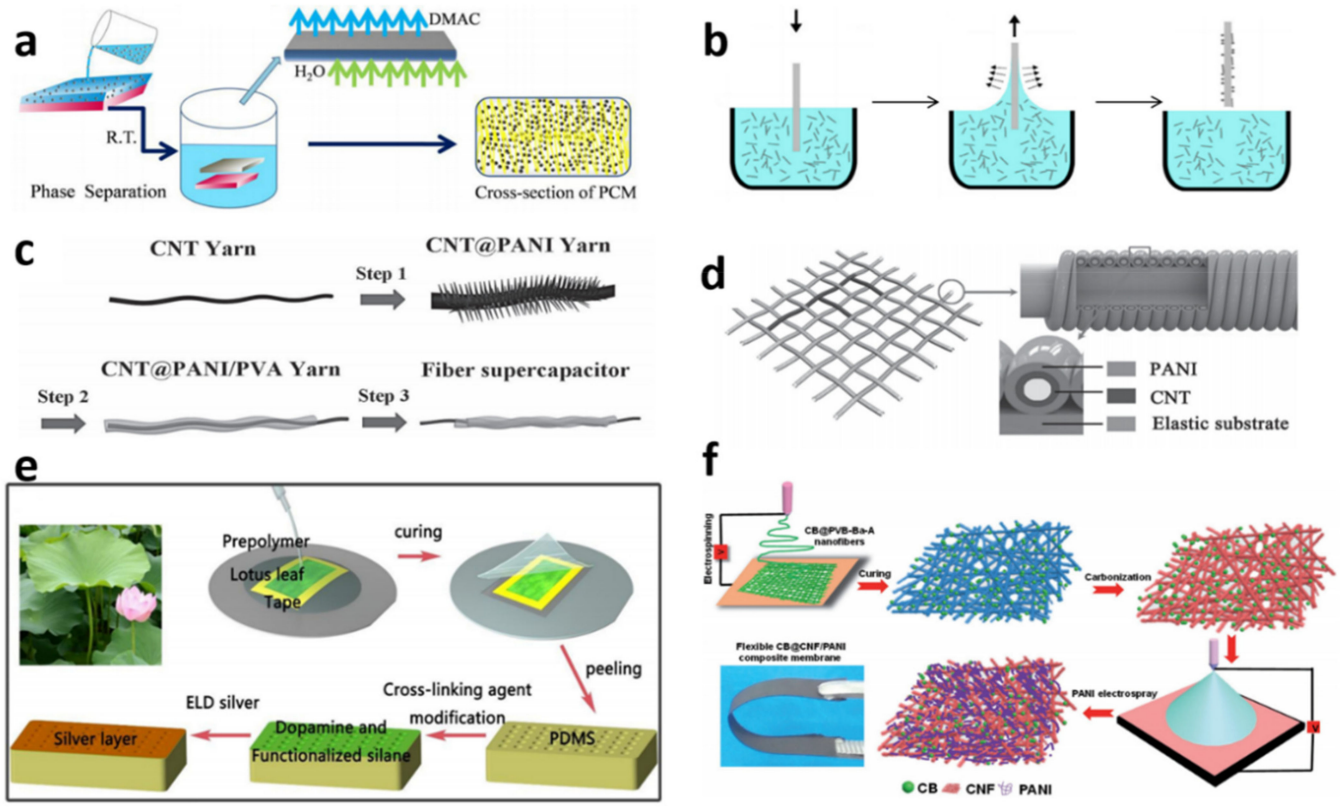

Figure 10. (**a**) Schematic illustration of PCM fabrication processes. Reprinted with permission from d. Copyright 2016 Journal of Power Sources. (**b**) The images illustrate the workflow of dip-coating. Reprinted with permission from [146]. Copyright 2012 Organic Electronics. (**c**) Schematics of the preparation procedures for the two-ply yarn supercapacitors. Reprinted with permission from [147]. Copyright 2013 Advanced Materials. (**d**) Schematic illustration of the structure wearable fiber-shaped supercapacitor. Reprinted with permission from [148]. Copyright 2014 Advanced Materials. (**e**) Schematic illustration of the fabrication process of the Cross-linking Intensified ELD-Ag/BEMF. Reprinted with permission from [149]. Copyright 2019 ACS Applied Materials & Interfaces. (**f**) Schematic illustration of synthesis pathway of flexible CB@CNF/PANI composite membrane. Reprinted with permission from [150]. Copyright 2017 Advanced Materials interfaces.

5.2. Chemical Oxidation Polymerization Method

This method is generally used to grow conductive materials on a suitable flexible matrix by a chemical in situ polymerization of monomers [151]. The key point of this method is to control the polymerization conditions of monomers and the synergistic forces between monomers and flexible matrices. Wang et al. [147] attached polyaniline nanowires formed by oxidative polymerization of aniline monomers to flexible carbon nanotubes as yarns (Figure 10c). Carbon nanotube yarns are used as both electrodes and high-toughness materials. In-situ polymerization of polyaniline nanowires on the surface of carbon nanotube yarns can further improve the energy storage capacity of supercapacitors. Similarly, Yu et al. [152] prepared flexible polyaniline/graphene composite electrodes using in situ oxidative polymerization of aniline.

5.3. Electrochemical Deposition

Electrochemical deposition is the most common among them, which mainly causes the monomers in the electrolyte to polymerize on the selected matrix under the action of an applied current to obtain a flexible electrode. This method is more suitable for conductive polymer and carbon-based materials. For example, Chen et al. [148] used rubber fibers as flexible substrates then attached carbon nanotubes to the flexible substrates, and finally

used the electrochemical deposition to deposit a layer of polyaniline on the surface of carbon nanotubes to obtain a flexible composite electrode (Figure 10d). Amade et al. [153] obtained a composite electrode with a thin MnO_2 layer wrapped in a thick carbon nanotube structure by electrochemical deposition of MnO_2 on carbon nanotubes, and obtained an ultra-high specific capacitance of 642 $f \cdot g^{-1}$ at a scanning speed of 10 $mV \cdot s^{-1}$. Hu and Lei [154] prepared nitrogen doped porous carbon fibers (NCF) using commercial carbon fibers by electrochemical deposition and calcination. The NCF was subjected to azide hydrothermal treatment to obtain a self-supporting NNCF flexible electrode.

5.4. Chemical/Physical Vapor Deposition

The "physical vapor deposition (PVD)" and "chemical vapor deposition (CVD)" method combines conductive material with flexible substrate, and the flexible stretchable electrodes obtained are mostly in membrane shape [155]. Compared with electrochemical deposition, vapor deposition is generally suitable for the preparation of some carbon-based materials such as graphene and carbon nanotubes. Niu et al. [156] deposited single-walled carbon nanotube thin films onto polydimethylsiloxanes by chemical vapor deposition, and then prepared stretchable flexible transducer devices using PVA/H_2SO_4 as gel electrolyte. Wu et al. [149] deposited silver on biomimetic elastomer microporous membranes by crosslinked enhanced ELD (electroless deposition) technology (Figure 10e). The bond strength between the PDMS substrate and the silver layer was significantly increased to 3.1 MPa by the low-cost upgraded ELD process and cross-linking enhancement. Although the materials prepared by physical vapor deposition have the advantages of small electrochemical impedance and uniform surface properties, their disadvantages are their expensive costs.

5.5. Electrospinning Technology

The material obtained by this technology has a high porosity, and its application in the electrochemical field will greatly increase the wettability between the material and the electrolyte, expand the accessible surface area of ions and help to improve the electrochemical performance of the device. Iqbal et al. [150] used the core-shell structure of carbon nanofibers/polyaniline composite as raw materials for spinning and obtained nanofibers composite membrane (Figure 10f) by "spinning-carbonization-spinning" three-step method. The supercapacitor prepared by this method also has good electrical properties. Zhang et al. [157] prepared a highly flexible transparent conductive electrode based on copper sulfide by electrostatic spinning and metal deposition. Nano-channel network (CuS NN) thin films have good chemical stability and significant mechanical stability.

5.6. Coating, Printing, and Spraying

This method has the advantages of low cost, easy scale production, and good substrate selectivity [158]. It can prepare a homogeneous suspension of active substances and coat them on flexible substrates by some means. For example, Xu et al. [159] used screen printing technology to print polyaniline/graphene composite slurry and apply it to the membrane electrodes of flexible supercapacitors (Figure 11a). Li et al. [160] prepared a foldable flexible supercapacitor by writing graphene slurry on a flexible substrate with a brush (Figure 11b). Lim et al. [161] were the first to obtain a flexible transparent conductive nano silver wire film, with an average square resistance of 38.7 $\Omega \cdot \square^{-1}$ and transmittance of 87.62%, on a PET substrate by brushing (Figure 11c).

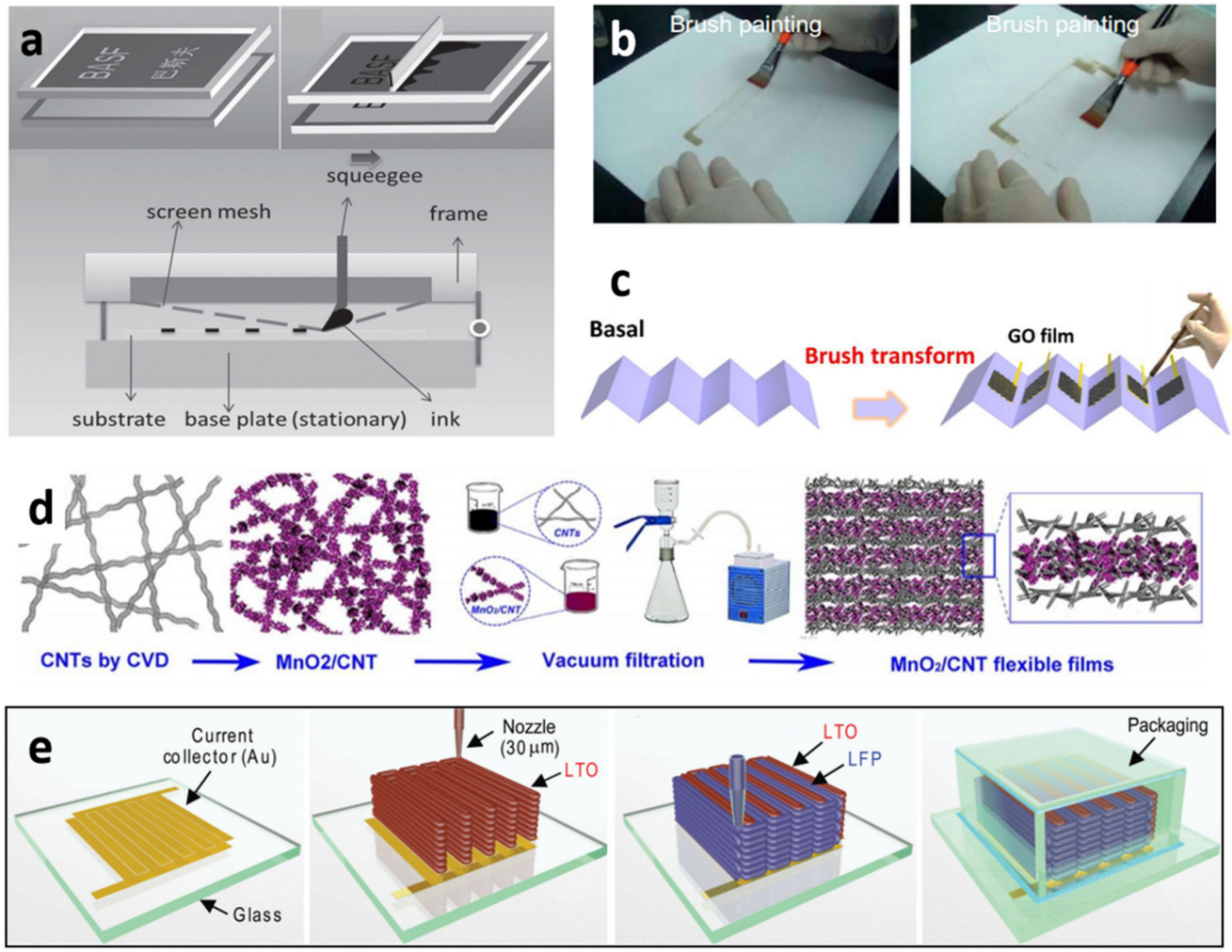

Figure 11. (**a**) Schematic illustration of the screen-printing process. Reprinted with permission from [159]. Copyright 2013 Advanced Energy Materials. (**b**) Brush-painting process of AgNWs on a PET substrate. Reprinted with permission from [160]. Copyright 2012 Solar Energy Materials & Solar Cells. (**c**) Schematic diagram showing the fabrication process for creating foldable supercapacitors. The rGO film can be directly written on a folded substrate and further reduced by illumination treatment. Reprinted with permission from [161]. Copyright 2017 Nano Energy. (**d**) Schematics of the stacked films' preparation process. Reprinted with permission from d. Copyright 2016 Advanced Energy Materials. (**e**) Schematic illustration of 3D interdigitated microbattery architectures (3D-IMA) fabricated on gold current collector by printing $Li_4Ti_5O_{12}$ (LTO) and $LiFePO_4$ (LFP) inks through 30μm nozzles, followed by sintering and packaging. Reprinted with permission from [162]. Copyright 2013 Advanced Materials.

5.7. 3D Printing

A new method of manufacturing parts directly from digital models by stacking layers of materials is called 3D printing [163]; It can use almost all types of materials, such as metals, ceramics, polymers, composites, and biomaterials. This method can enhance the specific capacity/capacitance, energy density and power density of energy storage equipment, and improve mechanical properties, especially high flexibility. Sun et al. [162] used 3D printing technology to manufacture a micron level micro battery with lithium iron phosphate as the positive electrode and lithium titanate as the negative electrode (Figure 11e). This micro battery has a high area specific capacity, which is the perfect combination of 3D printing technology and flexible energy storage devices. Li et al. produced a graphene composite aerogel microgrid electrode for flexible supercapacitors by 3D printing

technology [164]. The 3D printing graphene composite aerogel has the advantages of light weight, high conductivity, and excellent electrochemical performance. In particular, the flexible supercapacitors using 3D graphene composite aerogel electrodes show exceptional capacitance retention and power density. This work has extensive guiding significance for the manufacture of high-performance and integrated flexible energy storage devices.

5.8. Vacuum Suction

Compared with the previous methods, vacuum filtration is easier to operate, and the suspension of active substances deposits on the flexible substrates by gravity and vacuum pressure [165]. During the filtration process, the thin areas where the active substances accumulate are concentrated, and the velocity of the dispersed liquid is larger than that of the relatively thick areas, which results in a faster and thicker filtration rate in the thin areas, thus improving the uniformity of the membrane formation of the composite flexible electrodes. De et al. [166] fabricated a conductive network of nano-silver wires using the filter transfer method and attached it to the PET substrate. The resulting flexible transparent conductive film of nano-silver wires has 85% transparency, 13 $\Omega \cdot \square^{-1}$ average square resistance, and less than 2% resistance change after 1000 bending tests. Zeng et al. [167] prepared transparent conductive nano-silver wires thin films on glass substrates by filtration transfer method, then transferred the conductive network of nano-silver wires to polyvinyl Alcohol (PVA) substrates to obtain a flexible transparent conductive film with a transmittance of 87.5% at 550 nm and a mean square resistance of 63 $\Omega \cdot \square^{-1}$. The conductivity of the film can also be improved by alternating vacuum filtration, such as Wu et al. [168] using manganese dioxide (MnO_2)/carbon nanotube composite as active material and using alternating vacuum filtration method to prepare the self-supporting flexible membrane electrodes (Figure 11d).

6. Summary and Future Prospect

In this paper, the research progress of flexible stretchable electrodes and recent achievements in the application of flexible electronic devices have been introduced. The preparation of flexible stretchable electrodes with high linearity and high strain, and the study of sensing technology, provides the basic materials for the wide application of flexible electronic products. Flexible stretchable electrodes with high linearity and strain are characterized by flexibility, folding, twisting, compression, stretching, and arbitrary deformation while maintaining high conductivity and reliability. Its applications have penetrated many fields, such as industrial production, ocean exploration, environmental protection, medical diagnosis, bioengineering, space development, and smart home technology. For example, flexible stretchable solar cells, highly flexible actuators, stretchable supercapacitors, electronic skin, artificial muscles, and so on have been developed. High conductivity, stretchable and editable flexible conductive materials are the key materials to produce wearable electronic devices in the future, especially biomedical electronic devices that are perfectly compatible with human organs. The analysis and summary of this article have far-reaching significance for the development of these areas.

Although the research of flexible electrode technology has made great progress in the past few years, most of the preparation techniques are still in the preliminary stage. There are still some practical applications. In summary, there are three bottlenecks in the preparation of flexible stretchable electrode materials: (1) Low stability, including air stability and tensile stability; (2) Low reliability, which is mainly caused by the mismatch between the conductive substrate and the device interface; (3) Low integration, which is manifested by the low uniformity of large-size film-forming and device processing. To make a breakthrough in the future, the research on the combination of flexible electrodes with their application devices needs the new technology of integrating materials, electronics, machinery, and other disciplines and the cooperation of researchers from all disciplines.

Author Contributions: Writing—original draft preparation, W.H.; writing—review and editing, Q.L. and S.X.; supervision, Q.L., S.X., Y.S. and L.Q. All authors have read and agreed to the published version of the manuscript.

Funding: This work was supported by the National Natural Science Foundation of China (Nos. U2006218, 901203520, 51971029, 61871043, 62101055), BRICS STI Framework Programme by NSFC (No. 51861145309) and Qin Xin Talents Cultivation Program, Beijing Information Science & Technology University (QXTCP A202103).

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Data sharing is not applicable to this article.

Conflicts of Interest: The authors declare no conflict of interest.

References

1. Wang, C.; Xia, K.; Wang, H.; Liang, X.; Yin, Z.; Zhang, Y. Advanced carbon for flexible and wearable electronics. *Adv. Mater.* **2019**, *31*, e1801072. [CrossRef] [PubMed]
2. Shi, W.; Guo, Y.L.; Liu, Y.Q. When flexible organic field-effect transistors meet biomimetics: A prospective view of the internet of things. *Adv. Mater.* **2020**, *32*, 1901493. [CrossRef] [PubMed]
3. Shi, J.; Liu, S.; Zhang, L.; Yang, B.; Shu, L.; Yang, Y.; Ren, M.; Wang, Y.; Chen, J.; Chen, W.; et al. Smart textile-integrated microelectronic systems for wearable applications. *Adv. Mater.* **2020**, *32*, e1901958. [CrossRef] [PubMed]
4. Ma, Y.; Zhang, Y.; Cai, S.; Han, Z.; Liu, X.; Wang, F.; Cao, Y.; Wang, Z.; Li, H.; Chen, Y.; et al. Flexible hybrid electronics for digital healthcare. *Adv. Mater.* **2020**, *32*, e1902062. [CrossRef]
5. Li, P.; Zhang, Y.; Zheng, Z. Polymer-assisted metal deposition (PAMD) for flexible and wearable electronics: Principle, materials, printing, and devices. *Adv. Mater.* **2019**, *31*, e1902987. [CrossRef]
6. Haziri, V.; Phal, S.; Boily, J.-F.; Berisha, A.; Tesfalidet, S. Oxygen interactions with covalently grafted 2D nanometric carboxyphenyl thin films—An experimental and DFT study. *Coatings* **2022**, *12*, 49. [CrossRef]
7. Root, S.E.; Savagatrup, S.; Printz, A.; Rodriquez, D.; Lipomi, D.J. Mechanical properties of organic semiconductors for stretchable, highly flexible, and mechanically robust electronics. *Chem. Rev.* **2017**, *117*, 6467–6499. [CrossRef]
8. Kant, T.; Shrivas, K.; Ganesan, V.; Mahipal, Y.K.; Devi, R.; Deb, M.K.; Shankar, R. Flexible printed paper electrode with silver nano-ink for electrochemical applications. *Microchem. J.* **2020**, *155*, 104687. [CrossRef]
9. Liu, K.; Yao, Y.; Lv, T.; Li, H.; Li, N.; Chen, Z.; Qian, G.; Chen, T. Textile-like electrodes of seamless graphene/nanotubes for wearable and stretchable supercapacitors. *J. Power Sources* **2020**, *446*, 227355. [CrossRef]
10. Hu, R.; Wang, Y.; Zhao, J.; Jiang, R.; Zheng, J. Fabrication of stretchable multi-element composite for flexible solid-state electrochemical capacitor application. *Chem. Eng. J.* **2019**, *361*, 109–116. [CrossRef]
11. Tian, B.; Zheng, J.; Zhao, C.; Liu, C.; Su, C.; Tang, W.; Li, X.; Ning, G.-H. Correction: Carbonyl-based polyimide and polyquinoneimide for potassium-ion batteries. *J. Mater. Chem. A* **2019**, *7*, 12900. [CrossRef]
12. Zhang, Y.; Jiao, Y.; Liao, M.; Wang, B.; Peng, H. Carbon nanomaterials for flexible lithium ion batteries. *Carbon* **2017**, *124*, 79–88. [CrossRef]
13. Li, D.; Lai, W.-Y.; Zhang, Y.; Huang, W. Printable transparent conductive films for flexible electronics. *Adv. Mater.* **2018**, *30*, 1704738. [CrossRef] [PubMed]
14. Patel, M.; Seo, J.H.; Nguyen, T.T.; Kim, J. Active energy-controlling windows incorporating transparent photovoltaics and an integrated transparent heater. *Cell Rep. Phys. Sci.* **2021**, *2*, 100591. [CrossRef]
15. Bhatnagar, P.; Hong, J.; Patel, M.; Kim, J. Transparent photovoltaic skin for artificial thermoreceptor and nociceptor memory. *Nano Energy* **2021**, *91*, 106676. [CrossRef]
16. Wong, W.S.; Alberto, S. *Flexible Electronics: Materials and Applications*; Springer Science & Business Media: Berlin/Heidelberg, Germany, 2009.
17. Lipomi, D.J.; Vosgueritchian, M.; Tee, B.C.K.; Hellstrom, S.L.; Lee, J.A.; Fox, C.H.; Bao, Z. Skin-like pressure and strain sensors based on transparent elastic films of carbon nanotubes. *Nat. Nanotechnol.* **2011**, *6*, 788–792. [CrossRef]
18. Kang, S.-K.; Koo, J.; Lee, Y.K.; Rogers, J.A. Advanced materials and devices for bioresorbable electronics. *Accounts Chem. Res.* **2018**, *51*, 988–998. [CrossRef]
19. Chu, B.; Burnett, W.; Chung, J.W.; Bao, Z. Bring on the bodyNET. *Nature* **2017**, *549*, 328–330. [CrossRef]
20. Gao, W.; Emaminejad, S.; Nyein, H.Y.Y.; Challa, S.; Chen, K.; Peck, A.; Fahad, H.M.; Ota, H.; Shiraki, H.; Kiriya, D.; et al. Fully integrated wearable sensor arrays for multiplexed in situ perspiration analysis. *Nature* **2016**, *529*, 509–514. [CrossRef]
21. Lee, S.; Ha, G.; Wright, D.E.; Ma, Y.; Sen-Gupta, E.; Haubrich, N.R.; Branche, P.C.; Li, W.; Huppert, G.L.; Johnson, M.; et al. Highly flexible, wearable, and disposable cardiac biosensors for remote and ambulatory monitoring. *NPJ Digit. Med.* **2018**, *1*, 2. [CrossRef]

22. Liu, Y.; Norton, J.J.S.; Qazi, R.; Zou, Z.; Ammann, K.R.; Liu, H.; Yan, L.; Tran, P.L.; Jang, K.-I.; Lee, J.W.; et al. Epidermal mechano-acoustic sensing electronics for cardiovascular diagnostics and human-machine interfaces. *Sci. Adv.* **2016**, *2*, e1601185. [CrossRef] [PubMed]
23. Xu, B.; Akhtar, A.; Liu, Y.; Chen, H.; Yeo, W.-H.; Park, S.I.; Boyce, B.; Kim, H.; Yu, J.; Lai, H.-Y.; et al. An epidermal stimulation and sensing platform for sensorimotor prosthetic control, management of lower back exertion, and electrical muscle activation. *Adv. Mater.* **2016**, *28*, 4462–4471. [CrossRef] [PubMed]
24. Kim, D.-H.; Lu, N.; Ma, R.; Kim, Y.-S.; Kim, R.-H.; Wang, S.; Wu, J.; Won, S.M.; Tao, H.; Islam, A.; et al. Epidermal electronics. *Science* **2011**, *333*, 838–843. [CrossRef] [PubMed]
25. Li, H.; Xu, Y.; Li, X.; Chen, Y.; Jiang, Y.; Zhang, C.; Lu, B.; Wang, J.; Ma, Y.; Chen, Y.; et al. Epidermal inorganic optoelectronics for blood oxygen measurement. *Adv. Healthc. Mater.* **2017**, *6*, 1601013. [CrossRef]
26. Park, S.I.; Brenner, D.S.; Shin, G.; Morgan, C.D.; Copits, B.A.; Chung, H.U.; Pullen, M.Y.; Noh, K.N.; Davidson, S.; Oh, S.J.; et al. Soft, stretchable, fully implantable miniaturized optoelectronic systems for wireless optogenetics. *Nat. Biotechnol.* **2015**, *33*, 1280–1286. [CrossRef]
27. Feiner, R.; Dvir, T. Tissue–electronics interfaces: From implantable devices to engineered tissues. *Nat. Rev. Mater.* **2018**, *3*, 17076. [CrossRef]
28. Minev, I.R.; Musienko, P.; Hirsch, A.; Barraud, Q.; Wenger, N.; Moraud, E.M.; Gandar, J.; Capogrosso, M.; Milekovic, T.; Asboth, L.; et al. Electronic dura mater for long-term multimodal neural interfaces. *Science* **2015**, *347*, 159–163. [CrossRef]
29. "Electronic Plaster" Came Out? It Can Monitor Various Health Data of Human Body. Available online: https://zhuanlan.zhihu.com/p/351604777 (accessed on 2 April 2022).
30. The New App Only Needs to Take Photos of the Nail Bed to Accurately Judge the Symptoms of Anemia. Available online: https://www.sohu.com/a/279876829_100005926 (accessed on 2 April 2022).
31. Apple Watch Sales Exceeded 100 Million and Is Expected to Surpass MAC Computers. Available online: https://www.sohu.com/a/450746414_114822 (accessed on 2 April 2022).
32. From Trillion Market to One Bubble, Four Years History of Consumer Robots. Available online: https://www.sohu.com/a/226167819_460436 (accessed on 2 April 2022).
33. Liu, X. "Octopus Robot": Opening a New Era of Software Robot. Available online: http://www.chinainc.org.cn/show-14-104546-1.html (accessed on 2 April 2022).
34. Artificial Intelligence Is Expected to Be Used to Predict Tumor Lesions. Available online: https://www.sohu.com/a/253004726_100101359 (accessed on 2 April 2022).
35. Rubber Semiconductor: Bring Stretchable Electronic Devices Closer to Commercialization. Available online: https://m.sohu.com/a/293201739_427506 (accessed on 2 April 2022).
36. Wang, Y. An Exclusive Interview with Feng Guanping, Chairman of Shenwang New Material Technology Co., Ltd. Available online: https://www.cechoice.com/article/34160_3.html (accessed on 2 April 2022).
37. Chinese Academy of Sciences Has Developed a Large Area Full Fabric Pressure Sensor Array. Available online: https://www.ctn1986.com/index.php?c=content&a=show&id=75006 (accessed on 2 April 2022).
38. Interview with Cheng Huanyu, the First Person in Xianyang on the Forbes List. Available online: https://www.sohu.com/a/124592411_119659 (accessed on 2 April 2022).
39. Kim, D.-H.; Cha, G.D. Deformable inorganic semiconductor. *Nat. Mater.* **2018**, *17*, 388–389. [CrossRef]
40. Vaseem, M.; Mckerricher, G.; Shamim, A. robust design of a particle-free silver-organo-complex ink with high conductivity and inkjet stability for flexible electronics. *ACS Appl. Mater. Interfaces* **2015**, *8*, 177–186. [CrossRef]
41. Kou, P.; Yang, L.; Chang, C.; He, S. Improved flexible transparent conductive electrodes based on silver nanowire networks by a simple sunlight illumination approach. *Sci. Rep.* **2017**, *7*, 42052. [CrossRef]
42. Kumar, A.; Karadan, P.; Barshilia, H.C. Synthesis of silver nanowires towards the development of the ultrasensitive AgNWs/Si NPLs hybrid photodetector and flexible transparent conductor. *Mater. Sci. Semicond. Process.* **2018**, *75*, 239–246. [CrossRef]
43. Li, J.; Tao, Y.; Chen, S.; Li, H.; Chen, P.; Wei, M.-Z.; Wang, H.; Li, K.; Mazzeo, M.; Duan, Y. A flexible plasma-treated silver-nanowire electrode for organic light-emitting devices. *Sci. Rep.* **2017**, *7*, 16468. [CrossRef]
44. Magdassi, S.; Grouchko, M.; Berezin, O.; Kamyshny, A. Triggering the sintering of silver nanoparticles at room temperature. *ACS Nano* **2010**, *4*, 1943–1948. [CrossRef] [PubMed]
45. Nyholm, L.; Nyström, G.; Mihranyan, A.; Strømme, M. Toward flexible polymer and paper-based energy storage devices. *Adv. Mater.* **2011**, *23*, 3751–3769. [CrossRef] [PubMed]
46. Zhu, Y.; Yang, M.; Huang, Q.; Wang, D.; Yu, R.; Wang, J.; Zheng, Z.; Wang, D. V_2O_5 textile cathodes with high capacity and stability for flexible lithium-ion batteries. *Adv. Mater.* **2020**, *32*, 1906205. [CrossRef] [PubMed]
47. Trung, T.Q.; Lee, N.-E. Recent progress on stretchable electronic devices with intrinsically stretchable components. *Adv. Mater.* **2016**, *29*, 1603167. [CrossRef] [PubMed]
48. Rogers, J.A.; Someya, T.; Huang, Y. Materials and mechanics for stretchable electronics. *Science* **2010**, *327*, 1603–1607. [CrossRef] [PubMed]
49. Zhou, L.; Hu, Y.; Gao, H.; Gao, Y.; Zhu, W.; Zhan, L.; Liu, H.; Chen, Y.; Hu, K.; Wang, P.; et al. Moisture-assisted formation of high-quality silver nanowire transparent conductive films with low junction resistance. *Coatings* **2021**, *11*, 671. [CrossRef]

50. Hammock, M.L.; Chortos, A.; Tee, B.C.-K.; Tok, J.B.-H.; Bao, Z. 25th anniversary article: The evolution of electronic skin (E-skin): A brief history, design considerations, and recent progress. *Adv. Mater.* **2013**, *25*, 5997–6038. [CrossRef]
51. Magalhaes, B.; Engelhardt, S.; Molin, C.; Gebhardt, S.E.; Nielsch, K.; Hühne, R. Structural and electric properties of epitaxial $Na_{0.5}Bi_{0.5}TiO_3$-based thin films. *Coatings* **2021**, *11*, 651. [CrossRef]
52. Liu, Z.; Qian, Z.; Song, J.; Zhang, Y. Conducting and stretchable composites using sandwiched graphene-carbon nanotube hybrids and styrene-butadiene rubber. *Carbon* **2019**, *149*, 181–189. [CrossRef]
53. Sotelo, J.; Bonilla-Ríos, J.; García-Escobar, F.; Gordillo, J. Film growth of tetragonal SnO_2 on glass substrate by dip-coating technique for ethanol sensing applications. *Coatings* **2021**, *11*, 303. [CrossRef]
54. Zhu, C.; Liu, T.; Qian, F.; Han, T.Y.-J.; Duoss, E.B.; Kuntz, J.; Spadaccini, C.M.; Worsley, M.A.; Li, Y. Supercapacitors based on three-dimensional hierarchical graphene aerogels with periodic macropores. *Nano Lett.* **2016**, *16*, 3448–3456. [CrossRef]
55. Wang, Z.; Liu, X.; Shen, X.; Han, N.M.; Wu, Y.; Zheng, Q.; Jia, J.; Wang, N.; Kim, J. An ultralight graphene honeycomb sandwich for stretchable light-emitting displays. *Adv. Funct. Mater.* **2018**, *28*, 1707043. [CrossRef]
56. Sekitani, T.; Nakajima, H.; Maeda, H.; Fukushima, T.; Aida, T.; Hata, K.; Someya, T. Stretchable active-matrix organic light-emitting diode display using printable elastic conductors. *Nat. Mater.* **2009**, *8*, 494–499. [CrossRef] [PubMed]
57. Yu, K.J.; Kuzum, D.; Hwang, S.-W.; Kim, B.H.; Juul, H.; Kim, N.H.; Won, S.M.; Chiang, K.; Trumpis, M.; Richardson, A.G. Bioresorbable silicon electronics for transient spatiotemporal mapping of electrical activity from the cerebral cortex. *Nat. Mater.* **2016**, *15*, 782–791. [CrossRef]
58. Chen, T.; Peng, H.; Durstock, M.F.; Dai, L. High-performance transparent and stretchable all-solid supercapacitors based on highly aligned carbon nanotube sheets. *Sci. Rep.* **2015**, *4*, 3612. [CrossRef]
59. Xu, P.; Gu, T.; Cao, Z.; Wei, B.; Yu, J.; Li, F.; Byun, J.-H.; Lu, W.; Li, Q.; Chou, T.-W. Carbon nanotube fiber based stretchable wire-shaped supercapacitors. *Adv. Energy Mater.* **2013**, *4*, 1300759. [CrossRef]
60. Ren, J.; Zhang, Y.; Bai, W.; Chen, X.; Zhang, Z.; Fang, X.; Weng, W.; Wang, Y.; Peng, H. Elastic and wearable wire-shaped lithium-ion battery with high electrochemical performance. *Angew. Chem.* **2014**, *126*, 7998–8003. [CrossRef]
61. Zang, J.; Cao, C.; Feng, Y.; Liu, J.; Zhao, X. Stretchable and high-performance supercapacitors with crumpled graphene papers. *Sci. Rep.* **2015**, *4*, 6492. [CrossRef]
62. Lu, Z.; Foroughi, J.; Wang, C.; Long, H.; Wallace, G.G. Superelastic hybrid CNT/graphene fibers for wearable energy storage. *Adv. Energy Mater.* **2018**, *8*, 1702047. [CrossRef]
63. Lv, Z.; Luo, Y.; Tang, Y.; Wei, J.; Zhu, Z.; Zhou, X.; Li, W.; Zeng, Y.; Zhang, W.; Zhang, Y.; et al. Editable supercapacitors with customizable stretchability based on mechanically strengthened ultralong MnO_2 nanowire composite. *Adv. Mater.* **2018**, *30*, 1704531. [CrossRef] [PubMed]
64. Zhang, M.; Liu, Y.; Hu, M.; Wei, H.; Gao, Y. Spiral wire-type stretchable all-solid-state supercapacitors based on MnO_2/graphene/Ni wires. *Electrochim. Acta* **2017**, *256*, 44–51. [CrossRef]
65. Lv, T.; Yao, Y.; Li, N.; Chen, T. Highly stretchable supercapacitors based on aligned carbon nanotube/molybdenum disulfide composites. *Angew. Chem. Int. Ed.* **2016**, *55*, 9191–9195. [CrossRef]
66. Huang, J.; Li, D.; Zhao, M.; Mensah, A.; Lv, P.; Tian, X.; Huang, F.; Ke, H.; Wei, Q. Highly sensitive and stretchable CNT-bridged AgNP strain sensor based on TPU electrospun membrane for human motion detection. *Adv. Electron. Mater.* **2019**, *5*, 1900241. [CrossRef]
67. Li, Z.; Huang, T.; Gao, W.; Xu, Z.; Chang, D.; Zhang, C.; Gao, C. Hydrothermally activated graphene fiber fabrics for textile electrodes of supercapacitors. *ACS Nano* **2017**, *11*, 11056–11065. [CrossRef]
68. Xiong, Z.C.; Liao, W.H. Mechanically tough large-area hierarchical porous graphene films for high-performance flexible supercapacitor applications. *Adv. Mater.* **2015**, *27*, 4469–4475. [CrossRef]
69. Zhang, D.; Miao, M.; Niu, H.; Wei, Z. Core-spun carbon nanotube yarn supercapacitors for wearable electronic textiles. *ACS Nano* **2014**, *8*, 4571–4579. [CrossRef]
70. Souri, H.; Bhattacharyya, D. Highly Stretchable multifunctional wearable devices based on conductive cotton and wool fabrics. *ACS Appl. Mater. Interfaces* **2018**, *10*, 20845–20853. [CrossRef]
71. Wen, L.; Li, F.; Cheng, H.-M. Carbon nanotubes and graphene for flexible electrochemical energy storage: From materials to devices. *Adv. Mater.* **2016**, *28*, 4306–4337. [CrossRef]
72. Boutry, C.M.; Negre, M.; Jorda, M.; Vardoulis, O.; Chortos, A.; Khatib, O.; Bao, Z. A hierarchically patterned, bioinspired e-skin able to detect the direction of applied pressure for robotics. *Sci. Robot.* **2018**, *3*, eaau6914. [CrossRef] [PubMed]
73. Xiao, X.; Peng, X.; Jin, H.; Li, T.; Zhang, C.; Gao, B.; Hu, B.; Huo, K.; Zhou, J. Freestanding mesoporous VN/CNT hybrid electrodes for flexible all-solid-state supercapacitors. *Adv. Mater.* **2013**, *25*, 5091–5097. [CrossRef] [PubMed]
74. Niu, Z.; Luan, P.; Shao, Q.; Dong, H.; Li, J.; Chen, J.; Zhao, D.; Cai, L.; Zhou, W.; Chen, X.; et al. A "skeleton/skin" strategy for preparing ultrathin free-standing single-walled carbon nanotube/polyaniline films for high performance supercapacitor electrodes. *Energy Environ. Sci.* **2012**, *5*, 8726–8733. [CrossRef]
75. Xu, Z.; Liu, Z.; Sun, H.; Gao, C. Highly electrically conductive Ag-doped graphene fibers as stretchable conductors. *Adv. Mater.* **2013**, *25*, 3249–3253. [CrossRef]
76. Zhou, K.; Zhao, Y.; Sun, X.; Yuan, Z.; Zheng, G.; Dai, K.; Mi, L.; Pan, C.; Liu, C.; Shen, C. Ultra-stretchable triboelectric nanogenerator as high-sensitive and self-powered electronic skins for energy harvesting and tactile sensing. *Nano Energy* **2020**, *70*, 104546. [CrossRef]

77. Zhang, F.; Wu, S.; Peng, S.; Sha, Z.; Wang, C.H. Synergism of binary carbon nanofibres and graphene nanoplates in improving sensitivity and stability of stretchable strain sensors. *Compos. Sci. Technol.* **2019**, *172*, 7–16. [CrossRef]
78. Guo, K.; Wang, X.; Hu, L.; Zhai, T.; Li, H.; Yu, N. Highly Stretchable Waterproof Fiber Asymmetric Supercapacitors in an Integrated Structure. *ACS Appl. Mater. Interfaces* **2018**, *10*, 19820–19827. [CrossRef]
79. Zhang, Y.; Bai, W.; Cheng, X.; Ren, J.; Weng, W.; Chen, P.; Fang, X.; Zhang, Z.; Peng, H. Flexible and stretchable lithium-ion batteries and supercapacitors based on electrically conducting carbon nanotube fiber springs. *Angew. Chem. Int. Ed.* **2014**, *53*, 14564–14568. [CrossRef]
80. Tang, Q.; Chen, M.; Yang, C.; Wang, W.; Bao, H.; Wang, G. Enhancing the energy density of asymmetric stretchable supercapacitor based on wrinkled CNT@MnO_2 cathode and CNT@polypyrrole anode. *ACS Appl. Mater. Interfaces* **2015**, *7*, 15303–15313. [CrossRef]
81. Yun, T.G.; Hwang, B.I.; Kim, D.; Hyun, S.; Han, S.M. Polypyrrole–MnO_2-coated textile-based flexible-stretchable supercapacitor with high electrochemical and mechanical reliability. *ACS Appl. Mater. Interfaces* **2015**, *7*, 9228–9234. [CrossRef]
82. Hu, L.; Pasta, M.; La Mantia, F.; Cui, L.; Jeong, S.; Deshazer, H.D.; Choi, J.W.; Han, S.M.; Cui, Y. Stretchable, porous, and conductive energy textiles. *Nano Lett.* **2010**, *10*, 708–714. [CrossRef] [PubMed]
83. Kim, D.; Shin, G.; Kang, Y.J.; Kim, W.; Ha, J.S. Fabrication of a stretchable solid-state micro-supercapacitor array. *ACS Nano* **2013**, *7*, 7975–7982. [CrossRef] [PubMed]
84. Guo, F.M.; Xu, R.Q.; Cui, X.; Zhang, L.; Wang, K.L.; Yao, Y.W.; Wei, J.Q. High performance of stretchable carbon nanotube–polypyrrole fiber supercapacitors under dynamic deformation and temperature variation. *J. Mater. Chem. A* **2016**, *4*, 9311–9318. [CrossRef]
85. Yang, Y.; Sun, N.; Wen, Z.; Cheng, P.; Zheng, H.; Shao, H.; Xia, Y.; Chen, C.; Lan, H.; Xie, X.; et al. Liquid-metal-based super-stretchable and structure-designable triboelectric nanogenerator for wearable electronics. *ACS Nano* **2018**, *12*, 2027–2034. [CrossRef] [PubMed]
86. So, J.-H.; Koo, H.-J.; Dickey, M.D.; Velev, O.D. Ionic current rectification in soft-matter diodes with liquid-metal electrodes. *Adv. Funct. Mater.* **2012**, *22*, 625–631. [CrossRef]
87. Liu, Y.; Gao, M.; Mei, S.; Han, Y.; Liu, J. Ultra-compliant liquid metal electrodes with in-plane self-healing capability for dielectric elastomer actuators. *Appl. Phys. Lett.* **2013**, *103*, 064101. [CrossRef]
88. Gilshteyn, E.P.; Amanbayev, D.; Anisimov, A.S.; Kallio, T.; Nasibulin, A.G. All-nanotube stretchable supercapacitor with low equivalent series resistance. *Sci. Rep.* **2017**, *7*, 17449. [CrossRef]
89. Shi, J.; Wang, L.; Dai, Z.; Zhao, L.; Du, M.; Li, H.; Fang, Y. Multiscale hierarchical design of a flexible piezoresistive pressure sensor with high sensitivity and wide linearity range. *Small* **2018**, *14*, e1800819. [CrossRef]
90. Zhang, Y.; Sheehan, C.J.; Zhai, J.; Zou, G.; Luo, H.; Xiong, J.; Zhu, Y.; Jia, Q.X. Polymer-embedded carbon nanotube ribbons for stretchable conductors. *Adv. Mater.* **2010**, *22*, 3027–3031. [CrossRef]
91. Lin, J.; Peng, Z.; Liu, Y.; Ruiz-Zepeda, F.; Ye, R.; Samuel, E.L.G.; Yacaman, M.J.; Yakobson, B.I.; Tour, J.M. Laser-induced porous graphene films from commercial polymers. *Nat. Commun.* **2014**, *5*, 5714. [CrossRef]
92. Lamberti, A.; Clerici, F.; Fontana, M.; Scaltrito, L.A. Highly stretchable supercapacitor using laser-induced graphene electrodes onto elastomeric substrate. *Adv. Energy Mater.* **2016**, *6*, 1600050. [CrossRef]
93. Choi, T.Y.; Hwang, B.-U.; Kim, B.-Y.; Trung, T.Q.; Nam, Y.H.; Kim, D.-N.; Eom, K.; Lee, N.-E. Stretchable, transparent, and stretch-unresponsive capacitive touch sensor array with selectively patterned silver nanowires/reduced graphene oxide electrodes. *ACS Appl. Mater. Interfaces* **2017**, *9*, 18022–18030. [CrossRef] [PubMed]
94. Zhang, Z.; Wang, L.; Li, Y.; Wang, Y.; Zhang, J.; Guan, G.; Pan, Z.; Zheng, G.; Peng, H. Nitrogen-doped core-sheath carbon nanotube array for highly stretchable supercapacitor. *Adv. Energy Mater.* **2017**, *7*, 1601814. [CrossRef]
95. Li, M.; Chen, Q.; Zhan, H. Ultrathin manganese dioxide nanosheets grown on partially unzipped nitrogen-doped carbon nanotubes for high-performance asymmetric supercapacitors. *J. Alloy. Compd.* **2017**, *702*, 236–243. [CrossRef]
96. Wang, J.; Jiu, J.; Nogi, M.; Sugahara, T.; Nagao, S.; Koga, H.; He, P.; Suganuma, K. A highly sensitive and flexible pressure sensor with electrodes and elastomeric interlayer containing silver nanowires. *Nanoscale* **2015**, *7*, 2926–2932. [CrossRef]
97. Maeda, R.; Shinohara, Y.; Kawakami, H.; Isoda, Y.; Kanazawa, I.; Mitsuishi, M. The conducting fibrillar networks of a PEDOT:PSS hydrogel and an organogel prepared by the gel-film formation process. *Nanotechnol.* **2020**, *32*, 135403. [CrossRef] [PubMed]
98. Choong, C.-L.; Shim, M.-B.; Lee, B.-S.; Jeon, S.; Ko, D.-S.; Kang, T.-H.; Bae, J.; Lee, S.H.; Byun, K.-E.; Im, J.; et al. Highly stretchable resistive pressure sensors using a conductive elastomeric composite on a micropyramid array. *Adv. Mater.* **2014**, *26*, 3451–3458. [CrossRef]
99. Zhu, Y.; Li, N.; Lv, T.; Yao, Y.; Peng, H.; Shi, J.; Cao, S.; Chen, T. Ag-Doped PEDOT:PSS/CNT composites for thin-film all-solid-state supercapacitors with a stretchability of 480%. *J. Mater. Chem. A* **2018**, *6*, 941–947. [CrossRef]
100. Zhao, P.; Zhang, R.; Tong, Y.; Zhao, X.; Zhang, T.; Tang, Q.; Liu, Y. Strain-discriminable pressure/proximity sensing of transparent stretchable electronic skin based on PEDOT:PSS/SWCNT electrodes. *ACS Appl. Mater. Interfaces* **2020**, *12*, 55083–55093. [CrossRef]
101. Lee, J.H.; Jeong, Y.R.; Lee, G.; Jin, S.W.; Lee, Y.H.; Hong, S.Y.; Park, H.; Kim, J.W.; Lee, S.-S.; Ha, J.S. Highly conductive, stretchable, and transparent PEDOT:PSS electrodes fabricated with triblock copolymer additives and acid treatment. *ACS Appl. Mater. Interfaces* **2018**, *10*, 28027–28035. [CrossRef]
102. Ryu, S.; Lee, P.; Chou, J.B.; Xu, R.; Zhao, R.; Hart, A.J.; Kim, S.G. Fabrication of extremely elastic wearable strain sensor using aligned carbon nanotube fibers for monitoring human motion. *ACS Nano* **2015**, *9*, 5929–5936. [CrossRef] [PubMed]

103. Mai, H.; Mutlu, R.; Tawk, C.; Alici, G.; Sencadas, V. Ultra-stretchable MWCNT–Ecoflex piezoresistive sensors for human motion detection applications. *Compos. Sci. Technol.* **2019**, *173*, 118–124. [CrossRef]
104. Juyeon, Y.; Joonhyung, L.; Jaehyun, H. Stretchable supercapacitors based on carbon nanotubes-deposited rubber polymer nanofibers electrodes with high tolerance against strain. *Nanomaterials* **2018**, *8*, 541.
105. You, I.; Kim, B.; Park, J.; Koh, K.; Shin, S.; Jung, S.; Jeong, U. Stretchable E-skin apexcardiogram sensor. *Adv. Mater.* **2016**, *28*, 6359–6364. [CrossRef]
106. Gong, S.; Lai, D.T.; Wang, Y.; Yap, L.W.; Si, K.J.; Shi, Q.; Jason, N.N.; Sridhar, T.; Uddin, H.; Cheng, W. Tattoolike Polyaniline microparticle-doped gold nanowire patches as highly durable wearable sensors. *ACS Appl. Mater. Interfaces* **2015**, *7*, 19700–19708. [CrossRef] [PubMed]
107. Zhu, X.; Zhao, C.; Zhang, W.; Zhang, B.; Sun, M.; Chen, X.; Song, Y. Structural color control of CoFeB-coated nanoporous thin films. *Coatings* **2021**, *11*, 1123. [CrossRef]
108. Chen, Y.; Cai, K.; Liu, C.; Song, H.; Yang, X. High-performance and breathable polypyrrole coated air-laid paper for flexible all-solid-state supercapacitors. *Adv. Energy Mater.* **2017**, *7*, 1701247. [CrossRef]
109. Dong, L.; Xu, C.; Li, Y.; Pan, Z.; Liang, G.; Zhou, E.; Kang, F.; Yang, Q.-H. Breathable and wearable energy storage based on highly flexible paper electrodes. *Adv. Mater.* **2016**, *28*, 9313–9319. [CrossRef]
110. Xia, X.H.; Tu, J.P.; Zhang, Y.Q.; Mai, Y.J.; Wang, X.L.; Gu, C.D.; Zhao, X.B. Three-dimentional porous nano-Ni/$Co(OH)_2$ nanoflake composite film: A pseudocapacitive material with superior performance. *J. Phys. Chem. C* **2011**, *115*, 22662–22668. [CrossRef]
111. Li, Y.; Sheng, K.; Yuan, W.; Shi, G. A high-performance flexible fiber-shaped electrochemical capacitor based on electrochemically reduced graphene oxide. *Chem. Commun.* **2013**, *49*, 291–293. [CrossRef]
112. Yue, B.; Wang, C.; Ding, X.; Wallace, G. Polypyrrole coated nylon lycra fabric as stretchable electrode for supercapacitor applications. *Electrochim. Acta* **2012**, *68*, 18–24. [CrossRef]
113. Roh, E.; Hwang, B.-U.; Kim, D.; Kim, B.-Y.; Lee, N.-E. Stretchable, transparent, ultrasensitive, and patchable strain sensor for human-machine interfaces comprising a nanohybrid of carbon nanotubes and conductive elastomers. *ACS Nano* **2015**, *9*, 6252–6261. [CrossRef] [PubMed]
114. Qian, X.; Cai, Z.; Su, M.; Li, F.; Fang, W.; Li, Y.; Zhou, X.; Li, Q.; Feng, X.; Li, W.; et al. Printable skin-driven mechanoluminescence devices via nanodoped matrix modification. *Adv. Mater.* **2018**, *30*, e1800291. [CrossRef] [PubMed]
115. Ma, Z.; Huang, Q.; Xu, Q.; Zhuang, Q.; Zhao, X.; Yang, Y.; Qiu, H.; Yang, Z.; Wang, C.; Chai, Y.; et al. Permeable superelastic liquid-metal fibre mat enables biocompatible and monolithic stretchable electronics. *Nat. Mater.* **2021**, *20*, 859–868. [CrossRef]
116. Dong, R.; Wang, L.; Hang, C.; Chen, Z.; Liu, X.; Zhong, L.; Qi, J.; Huang, Y.; Liu, S.; Wang, L.; et al. Printed stretchable liquid metal electrode arrays for in vivo neural recording. *Small* **2021**, *17*, 2006612. [CrossRef] [PubMed]
117. He, W.; Wang, C.; Wang, H.; Jian, M.; Lu, W.; Liang, X.; Zhang, X.; Yang, F.; Zhang, Y. Integrated textile sensor patch for real-time and multiplex sweat analysis. *Sci. Adv.* **2019**, *5*, eaax0649. [CrossRef] [PubMed]
118. Yu, J.L.; Lu, W.B.; Smith, J.P.; Booksh, K.S.; Meng, L.H.; Huang, Y.D.; Li, Q.W.; Byun, J.H.; Oh, Y.; Yan, Y.S.; et al. A high performance stretchable asymmetric fiber-shaped supercapacitor with a core-sheath helical structure. *Adv. Energy Mater.* **2017**, *7*, 1600976. [CrossRef]
119. Chu, X.; Zhang, H.; Su, H.; Liu, F.; Gu, B.; Huang, H.; Zhang, H.; Deng, W.; Zheng, X.; Yang, W. A novel stretchable supercapacitor electrode with high linear capacitance. *Chem. Eng. J.* **2018**, *349*, 168–175. [CrossRef]
120. Lv, J.; Jeerapan, I.; Tehrani, F.; Yin, L.; Silva-Lopez, C.A.; Jang, J.-H.; Joshuia, D.; Shah, R.; Liang, Y.; Xie, L.; et al. Sweat-based wearable energy harvesting-storage hybrid textile devices. *Energy Environ. Sci.* **2018**, *11*, 3431–3442. [CrossRef]
121. Yang, Y.; Huang, Q.Y.; Niu, L.Y.; Wang, D.R.; Yan, C.; She, Y.Y.; Zheng, Z.J. Waterproof, Ultrahigh areal-capacitance, wearable supercapacitor fabrics. *Adv. Mater.* **2017**, *29*, 1606679. [CrossRef]
122. Bhagavatheswaran, E.S.; Parsekar, M.; Das, A.; Le, H.H.; Wiessner, S.; Stöckelhuber, K.W.; Schmaucks, G.; Heinrich, G. Construction of an interconnected nanostructured carbon black network: Development of highly stretchable and robust elastomeric conductors. *J. Phys. Chem. C* **2015**, *119*, 21723–21731. [CrossRef]
123. Ge, J.; Yao, H.-B.; Wang, X.; Ye, Y.-D.; Wang, J.-L.; Wu, Z.-Y.; Liu, J.-W.; Fan, F.-J.; Gao, H.-L.; Zhang, C.-L.; et al. Stretchable conductors based on silver nanowires: Improved performance through a binary network design. *Angew. Chem. Int. Ed.* **2013**, *52*, 1654–1659. [CrossRef] [PubMed]
124. Cheng, Y.; Wang, R.; Zhai, H.; Sun, J. Stretchable electronic skin based on silver nanowire composite fiber electrodes for sensing pressure, proximity, and multidirectional strain. *Nanoscale* **2017**, *9*, 3834–3842. [CrossRef] [PubMed]
125. Li, J.; Cao, P.; Tan, Y.; Zhang, L. Effect of the addition of TEOS on the SiC fibers fabricated by electrospinning. *J. Adv. Ceram.* **2013**, *2*, 242–245. [CrossRef]
126. Chen, Z.; Sun, X.; Shang, Y.; Xiong, K.; Xu, Z.; Guo, R.; Cai, S.; Zheng, C. Dense ceramics with complex shape fabricated by 3D printing: A review. *J. Adv. Ceram.* **2021**, *10*, 195–218. [CrossRef]
127. Kim, J.S.; Lee, S.C.; Hwang, J.; Lee, E.; Cho, K.; Kim, S.J.; Kim, D.H.; Lee, W.H. Enhanced sensitivity of iontronic graphene tactile sensors facilitated by spreading of ionic liquid pinned on graphene grid. *Adv. Funct. Mater.* **2020**, *30*, 1908993. [CrossRef]
128. Amjadi, M.; Pichitpajongkit, A.; Lee, S.; Ryu, S.; Park, I. Highly stretchable and sensitive strain sensor based on silver nanowire–elastomer nanocomposite. *ACS Nano* **2014**, *8*, 5154–5163. [CrossRef]
129. Dong, W.; Kong, L.; Kang, L.; Ran, F. Flexibility and microminiaturization of electrode materials and devices for supercapacitor. *Mater. Rep.* **2018**, *32*, 2912–2919.

130. Zhang, Q.; Zhou, K.; Lei, J.; Hu, W. Nitrogen dual-doped porous carbon fiber: A binder-free and high-performance flexible anode for lithium ion batteries. *Appl. Surf. Sci.* **2019**, *467–468*, 992–999. [CrossRef]
131. Qi, R.; Nie, J.; Liu, M.; Xia, M.; Lu, X. Stretchable V_2O_5/PEDOT supercapacitors: A modular fabrication process and charging with triboelectric nanogenerators. *Nanoscale* **2018**, *10*, 7719–7725. [CrossRef]
132. Dong, K.; Wang, Y.-C.; Deng, J.; Dai, Y.; Zhang, S.L.; Zou, H.; Gu, B.; Sun, B.; Wang, Z.L. A highly stretchable and washable all-yarn-based self-charging knitting power textile composed of fiber triboelectric nanogenerators and supercapacitors. *ACS Nano* **2017**, *11*, 9490–9499. [CrossRef] [PubMed]
133. Zhao, X.; Ran, F.; Shen, K.; Yang, Y.; Wu, J.; Niu, X.; Kong, L.; Kang, L.; Chen, S. Facile fabrication of ultrathin hybrid membrane for highly flexible supercapacitors via in-situ phase separation of polyethersulfone. *J. Power Sources* **2016**, *329*, 104–114. [CrossRef]
134. Durham, J.W.; Zhu, Y. Fabrication of functional nanowire devices on unconventional substrates using strain-release assembly. *ACS Appl. Mater. Interfaces* **2013**, *5*, 256–261. [CrossRef] [PubMed]
135. Xie, Y.; Liu, Y.; Zhao, Y.; Tsang, Y.H.; Lau, S.P.; Huang, H.; Chai, Y. Stretchable all-solid-state supercapacitor with wavy shaped polyaniline/graphene electrode. *J. Mater. Chem. A* **2014**, *2*, 9142–9149. [CrossRef]
136. Ren, D.; Dong, L.; Wang, J.; Ma, X.; Xu, C.; Kang, F. Facile Preparation of high-performance stretchable fiber-like electrodes and supercapacitors. *ChemistrySelect* **2018**, *3*, 4179–4184. [CrossRef]
137. Zhang, N.; Zhou, W.; Zhang, Q.; Luan, P.; Cai, L.; Yang, F.; Zhang, X.; Fan, Q.; Zhou, W.; Xiao, Z.; et al. Biaxially stretchable supercapacitors based on the buckled hybrid fiber electrode array. *Nanoscale* **2015**, *7*, 12492–12497. [CrossRef]
138. He, S.; Qiu, L.; Wang, L.; Cao, J.; Xie, S.; Gao, Q.; Zhang, Z.; Zhang, J.; Wang, B.; Peng, H. A three-dimensionally stretchable high performance supercapacitor. *J. Mater. Chem. A* **2016**, *4*, 14968–14973. [CrossRef]
139. Lee, Y.; Chae, S.; Park, H.; Kim, J.; Jeong, S.H. Stretchable and transparent supercapacitors based on extremely long MnO_2/Au nanofiber networks. *Chem. Eng. J.* **2020**, *382*, 122798. [CrossRef]
140. Kim, A.; Won, Y.; Woo, K.; Jeong, S.; Moon, J. All-solution-processed indium-free transparent composite electrodes based on Ag nanowire and metal oxide for thin-film solar cells. *Adv. Funct. Mater.* **2014**, *24*, 2462–2471. [CrossRef]
141. Yan, X.; Ma, J.; Xu, H.; Wang, C.; Liu, Y. Fabrication of silver nanowires and metal oxide composite transparent electrodes and their application in UV light-emitting diodes. *J. Phys. D Appl. Phys.* **2016**, *49*, 325103. [CrossRef]
142. Pu, J.; Wang, X.; Xu, R.; Komvopoulos, K. Highly stretchable microsupercapacitor arrays with honeycomb structures for integrated wearable electronic systems. *ACS Nano* **2016**, *10*, 9306–9315. [CrossRef] [PubMed]
143. Lv, Z.; Tang, Y.; Zhu, Z.; Wei, J.; Li, W.; Xia, H.; Jiang, Y.; Liu, Z.; Luo, Y.; Ge, X.; et al. Honeycomb-lantern-inspired 3D stretchable supercapacitors with enhanced specific areal capacitance. *Adv. Mater.* **2018**, *30*, e1805468. [CrossRef]
144. Wang, X.; Zhang, Y.; Zhang, X.; Huo, Z.; Li, X.; Que, M.; Peng, Z.; Wang, H.; Pan, C. A Highly stretchable transparent self-powered triboelectric tactile sensor with metallized nanofibers for wearable electronics. *Adv. Mater.* **2018**, *30*, e1706738. [CrossRef] [PubMed]
145. Zhou, G.; Kim, N.-R.; Chun, S.-E.; Lee, W.; Um, M.-K.; Chou, T.-W.; Islam, M.; Byun, J.-H.; Oh, Y. Highly porous and easy shapeable poly-dopamine derived graphene-coated single walled carbon nanotube aerogels for stretchable wire-type supercapacitors. *Carbon* **2018**, *130*, 137–144. [CrossRef]
146. Sachse, C.; Müller-Meskamp, L.; Bormann, L.; Kim, Y.H.; Lehnert, F.; Philipp, A.; Beyer, B.; Leo, K. Transparent, dip-coated silver nanowire electrodes for small molecule organic solar cells. *Org. Electron.* **2013**, *14*, 143–148. [CrossRef]
147. Huang, Y.; Tao, J.; Meng, W.; Zhu, M.; Fu, Y.; Gao, Y.; Zhi, C. Super-high rate stretchable polypyrrole-based supercapacitors with excellent cycling stability. *Nano Energy* **2015**, *11*, 518–525. [CrossRef]
148. Wang, K.; Meng, Q.; Zhang, Y.; Wei, Z.; Miao, M. High-performance two-ply yarn supercapacitors based on carbon nanotubes and polyaniline nanowire arrays. *Adv. Mater.* **2013**, *25*, 1494–1498. [CrossRef]
149. Yu, H.; Ge, X.; Bulin, C.; Xing, R.; Li, R.; Xin, G.; Zhang, B. Facile fabrication and energy storage analysis of graphene/PANI paper electrodes for supercapacitor application. *Electrochim. Acta* **2017**, *253*, 239–247. [CrossRef]
150. Chen, X.; Lin, H.; Deng, J.; Zhang, Y.; Sun, X.; Chen, P.; Fang, X.; Zhang, Z.; Guan, G.; Peng, H. Electrochromic fiber-shaped supercapacitors. *Adv. Mater.* **2014**, *26*, 8126–8132. [CrossRef]
151. Amade, R.; Jover, E.; Caglar, B.; Mutlu, T.; Bertran, E. Optimization of MnO_2/vertically aligned carbon nanotube composite for supercapacitor application. *J. Power Sources* **2011**, *196*, 5779–5783. [CrossRef]
152. Chen, X.; Lin, H.; Chen, P.; Guan, G.; Deng, J.; Peng, H. Smart, stretchable supercapacitors. *Adv. Mater.* **2014**, *26*, 4444–4449. [CrossRef] [PubMed]
153. Cheon, G.; Duerloo, K.-A.N.; Sendek, A.D.; Porter, C.; Chen, Y.; Reed, E.J. Data mining for new two- and one-dimensional weakly bonded solids and lattice-commensurate heterostructures. *Nano Lett.* **2017**, *17*, 1915–1923. [CrossRef] [PubMed]
154. Niu, Z.; Dong, H.; Zhu, B. Highly stretchable, integrated supercapacitors based on single-walled carbon nanotube films with continuo us reticulate architecture. *Adv. Mater.* **2013**, *25*, 1058–1064. [CrossRef] [PubMed]
155. Wu, C.; Tang, X.; Gan, L.; Li, W.; Zhang, J.; Wang, H.; Qin, Z.; Zhou, T.; Huang, J.; Xie, C.; et al. High-adhesion stretchable electrode via cross-linking intensified electroless deposition on a biomimetic elastomeric micropore film. *ACS Appl. Mater. Interfaces* **2019**, *11*, 20535–20544. [CrossRef]
156. Iqbal, N.; Wang, X.; Babar, A.A.; Yan, J.; Yu, J.; Park, S.-J.; Ding, B. Polyaniline enriched flexible carbon nanofibers with core-shell structure for high-performance wearable supercapacitors. *Adv. Mater. Interfaces* **2017**, *4*, 1700855. [CrossRef]

157. Zhang, X.; Guo, W.; Gao, G.; Que, M.; Pan, C.; Wang, Z.L. CuS nano through-networks for highly stable transparent conducting electrodes. *J. Phys. Chem. C* **2016**, *4*, 4733–4739.
158. Fraser, D.B.; Cook, H.D. High conductive transparent films of sputtered $In_{2-x}Sn_xO_{3-y}$. *J. Electrochem. Soc.* **1972**, *119*, 1368–1374. [CrossRef]
159. Xu, Y.; Schwab, M.G.; Strudwick, A.J.; Hennig, I.; Feng, X.; Wu, Z.; Müllen, K. Screen-printable thin film supercapacitor device utilizing graphene/polyaniline inks. *Adv. Energy Mater.* **2013**, *3*, 1035–1040. [CrossRef]
160. Li, J.; Shao, Y.; Shi, Q. Calligraphy-inspired brush written foldable supercapacitors. *Nano Energy* **2017**, *38*, 428–437. [CrossRef]
161. Lim, J.-W.; Cho, D.-Y.; Kim, J.-; Na, S.-I.; Kim, H.-K. Simple brush-painting of flexible and transparent Ag nanowire network electrodes as an alternative ITO anode for cost-efficient flexible organic solar cells. *Sol. Energy Mater. Sol. Cells* **2012**, *107*, 348–354. [CrossRef]
162. Sun, K.; Wei, T.-S.; Ahn, B.Y.; Seo, J.Y.; Dillon, S.J.; Lewis, J.A. 3D printing of interdigitated Li-ion microbattery architectures. *Adv. Mater.* **2013**, *25*, 4539–4543. [CrossRef] [PubMed]
163. Kang, M.-G.; Park, H.J.; Ahn, S.H.; Xu, T.; Guo, L.J. Toward low-cost, high-efficiency, and scalable organic solar cells with transparent metal electrode and improved domain morphology. *IEEE J. Sel. Top. Quantum Electron.* **2010**, *16*, 1807–1820. [CrossRef]
164. Liu, L.; Tian, Q.; Yao, W.; Li, M.; Li, Y.; Wu, W. All-printed Ultra flexible and stretchable asymmetric in-plane solid-state supercapacitors (ASCs) for wearable electronics. *J. Power Sources* **2018**, *397*, 59–67. [CrossRef]
165. De, S.; Higgins, T.M.; Lyons, P.E.; Doherty, E.M.; Nirmalraj, P.N.; Blau, W.; Boland, J.; Coleman, J. Silver Nanowire networks as flexible, transparent, conducting films: Extremely high DC to optical conductivity ratios. *ACS Nano* **2009**, *3*, 1767–1774. [CrossRef] [PubMed]
166. Zeng, X.-Y.; Zhang, Q.-K.; Yu, R.-M.; Lu, C.-Z. A new transparent conductor: Silver nanowire film buried at the surface of a transparent polymer. *Adv. Mater.* **2010**, *22*, 4484–4488. [CrossRef] [PubMed]
167. Wu, P.; Cheng, S.; Yang, L. Synthesis and characterization of self-standing and highly flexible delta-MnO_2@CNTs/CNTs composite films for direct use of supercapacitor electrodes. *ACS Appl. Mater. Interfaces* **2016**, *8*, 23721–23728. [CrossRef]
168. Guo, K.; Zhang, G.; Long, Y.; Ning, H.; Xu, Z.; Qiu, T.; Luo, D.; Li, M.; Yao, R.; Peng, J. Modifying precursor solutions to obtain screen-printable inks for tungsten oxides electrochromic film preparation. *Coatings* **2021**, *11*, 872. [CrossRef]