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Recent Progress on Stretchable Supercapacitors

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Abstract

Stretchable supercapacitors can function in versatile environments under multiple mechanical deformations including stretching, bending, twisting and compressing, which has implications for powering wearable electronics and implantable biomedical devices, where adaptability and compliant mechanics are required. This review covers the recent advances in the field of stretchable supercapacitors with a focus on materials and their design principles. Firstly, we discuss material aspects on stretchable supercapacitor electrode composition, which includes carbon-based materials, metal-based materials, and conductive polymers. Then, we describe novel strategies to construct 1D, 2D and 3D supercapacitors, highlighting their stretchability and electrochemical performances. Furthermore, we cover extra add-on functionalities that made stretchable supercapacitors ultrathin, transparent or self-healable, as well as their integration with other energy conversion devices. Finally, we discuss the challenges and opportunities in the future developments and optimization of stretchable supercapacitors.

1. Introduction

Wearable electronics is a rising industry with its enormous potential to revolutionize the way that human interacts and communicates with environments, in context of "big health" and "big data".[1-11] Thus, developing comparably wearable energy devices is increasingly crucial for powering those wearable electronics, which has experienced rapid growth over the past several years.[12-15] In order to assure the sustainable operation of the whole wearable electronic system, the corresponding power sources have to deliver stable and persistent electricity output as well as maintain their satisfactory performances under all kinds of deforming conditions such as bending, twisting and stretching, which requires the energy devices to be reasonably flexible and stretchable that could be assembled or attached on clothes or human body. Among various energy devices, stretchable supercapacitors hold great promise as the main power source for wearable electronics, not only owing to their superior power density, low cost, fast charging/discharging rate, long calendar life and simple construction that the conventional supercapacitors possess, the stretchability offers them the feasibility to work ideally under different environments as well as to be combined conveniently with other units to build the integrate wearable electronics system.[16] To date, research efforts have been devoted mostly to three aspects:

- The design and establishment of stretchable electrodes, current collectors and electro-active materials, while the versatile combination of composite materials and designing strategies results in significant differences in the obtained mechanical stretchability and electrochemical performances.
- The construction of the whole stretchable supercapacitor system including the assembly of electrodes, separator and electrolyte, based on different configuration types such as 1D linear supercapacitor, 2D planar supercapacitor and 3D stereo supercapacitor, in order to ensure the stable operation under multiple stretched states (uniaxial strain, biaxial strain or even 3D stretching strain) and prove some practical wearable applications, such as being attached to human skin or sewn into textiles.
- The other multi-functionalities such as lightweight, ultrathinness or transparency that have been added in order to realize the dream of the skin-conformable or invisible stretchable supercapacitor system, as well as device-level integration with other wearable electronics such as energy conversion devices. Those are also the ultimate goals for the stretchable energy devices.

To date, substantial achievements have been made in the first two aspects while the third one is still on its initial state that requires further improvements in the future.

This review aims to provide a comprehensive coverage of the progress made in the field of stretchable supercapacitors over the past several years. We skip the basic information such as the structures and electrochemical mechanism for supercapacitor, which may be found in the recent excellent reviews,[17-20] instead, we emphasize on the stretchable electrodes designing technologies along with the achieved mechanical stretchability, electrochemical performances, practical applications and multi-functionalities. Along with **Scheme 1** of the overall progressive research developments on stretchable supercapacitor, we first summarize the material aspects regarding substrates, current collectors and electro-active materials together for stretchable electrode. Then, the detailed stretchable electrodes designing strategies classified as 1D linear, 2D planar and 3D stereo along with the accomplished mechanical stretchability, related electrochemical performances and practical applications are described. Next, we highlight and discuss several representative examples which endow stretchable supercapacitors with multi-functionalities such as ultrathinness, transparency and self-healing properties, as well as the state-of-the-art attempts to integrate stretchable supercapacitors with other wearable electronics such as energy harvesters or strain sensors. Finally, the future directions and challenges for stretchable supercapacitors in real-world applications are discussed.

2. Electrode Material Considerations for Stretchable Supercapacitors

As the most important part of supercapacitors, electrode is responsible for the overall performances including electrochemical/mechanical stability, capacitance, charge-discharge efficiency as well as power/energy density. The first priority to achieve stretchable supercapacitor is to build stretchable electrode, which needs rational material selection and novel structure designs. In general, the substrates, current collectors and electro-active materials together constitute the overall electrode design for stretchable supercapacitors. The substrates for most stretchable electrodes at present are mainly elastomeric polymers with intrinsic stretchability such as PDMS (polydimethylsiloxane), PU (polyurethane), Ecoflex (silicone rubber) or SEBS (styrene-(ethylene-butylene)-styrene) that can be directly made into thin films or fibers for different types of stretchable electrodes. Other substrates include textiles, carbon fibers or cellulose papers which are less stretchable or intrinsically non-stretchable at all, but can be also made stretchable through extrinsic structural designs.

As for current collectors, metals such as gold, silver, platinum or copper are most suitable candidates as they are highly conductive, but the rigid nature of bulk metal materials seriously limit their application in designing stretchable electrodes because of the loss of conductivity even under small strains. In this context, nanostructured metal materials can circumvent the limitation by forming serpentine structures or percolation networks that are either on or embedded into polymer matrix.[21-23] Similar challenges are present for bulk carbon materials, hence, nanocarbon materials such as carbon nanotube (CNT) and graphene have been intensively researched for stretchable current collectors for supercapacitors.[24] It is commonly believed that noble materials such as gold may be costly, preventing them from real-world application. Nevertheless, it is not entirely true considering merits, such as small amount needed for biomedical applications, low-cost material processing and biocompatibility. In contrast, strong acids used for dissolving CNTs and graphene may involve nasty chemical processing with high cost.

With regards to electro-active materials, numerous capacitive materials have been studied for supercapacitors to date, such as carbon-based materials including CNT, graphene, graphite and other activated carbons providing electric double-layer capacitance (EDLC) or other metal compounds (such as metal oxides, metal sulfides or metal hydroxide) and conductive polymers with redox reactions to build pseudocapacitors.[25,26] Below we summarize the materials in detail for stretchable electrodes, classified into carbon-based material, metal-based material and conductive polymers.

2.1 Carbon-based materials

Carbon-based materials such as CNTs, graphene and their composites with different forms are the most commonly-used materials in stretchable electrode designs, which not only serve as electro-active materials but can also serve as current collectors in many stretchable supercapacitor systems. They have advantages such as large surface area, low density, good mechanical properties, and high stability.[27] For instance, CNTs have shown great superiority as stretchable electrode materials and have been widely used in many stretchable designs. As shown in **Figure 1**a, researchers transferred CNT film onto pre-strained PDMS film and buckled/wavy structures were formed after releasing the strain (**Figure 1**b).[28] This kind of buckled/wavy structure could render the electrodes with certain stretchability. The as-build stretchable supercapacitor could withstand 120% of strain with no significant deviation in electrochemical performance, with specific capacitance of 48 $F\cdot g^{-1}$. Further advance in specific capacity could be achieved if conductive polymer such as PANI (polyaniline) was electro-deposited on the CNT film as shown in **Figure 1**c.[29] With stretchability up to 100%, the specific capacity could reach as high as 308.4 F/g. CNT fibers were also utilized as stretchable electrodes. **Figure 1**d shows that two twisted CNT fibers were glued on a PDMS-coated spandex fiber under 100% pre-strain, which then were used to fabricate stretchable wire-shaped supercapacitors by releasing the strain.[30] Another way to effectively improve the stretchability as well as the electrochemical performance is doping other elements so as to add extra pseudo-capacity. Recently, nitrogen (N)-doped core-sheath carbon nanotube (NCNT) arrays were developed as an elastic electrode which exhibited 400% of stretchability, high areal specific capacity of 31.1 $mF\cdot cm^{-2}$, as well as superior mechanical and electrochemical stability (**Figure 1**e and f).[31]

Graphene has also been widely used in the fabrication of stretchable electrodes. Highly stretchable multilayer graphene sheets were synthesized on wrinkle-structured copper foil and transferred to PDMS substrate, as show in **Figure 2**a.[32] The as-prepared graphene sheets served both as active materials and current collectors, achieving 40% of stretchability as well as high optical transparency. Similar work was carried out in which four-layer buckled graphene films were synthesized on elastomeric PDMS substrates (**Figure 2**b).[33] In addition, graphene fiber (GF) was also used in stretchable electrodes. For instance, all graphene core-sheath fibers, denoted as 3D GF@3D-G, in which a core of GF was covered with a sheath of 3D porous network-like graphene framework, were fabricated as shown in **Figure 2**c and d.[34] Two 3D GF@3D-G fibers were twisted together with polyelectrolyte and wrapped around a glass rod in order to build spring-like stretchable supercapacitor, with stretchability up to 200% and areal specific capacity of 1.2 mF/cm^2.

2.2 Metal-based materials

Metal-based materials include pure conductive metals (gold, silver or copper) that function as current collectors and metal compounds that have redox reaction during electrochemical process. For current collectors nowadays, metal materials have been extensively used in many practical miniaturized electronic circuits owing to their high conductivity. In order to make bulk metal materials flexible or even stretchable, researchers have deposited nanostructured metal such as nanowires or nanoparticles with engineered layout on stretchable substrates or embedded them in polymer matrix to build the flexible/stretchable conductive film, which have been widely utilized in many wearable electronics.[21-23] In particular, our group have demonstrated that ultrathin gold nanowires (~2 nm thick, **Figure 3**a) could self-assemble into highly conductive and flexible electrodes with nanomesh microstructure (**Figure 3**b).[35] The formation of such nanomesh electrodes is due to partial oleylamine ligand removal during ageing or annealing process, which led to the configurational change from aligned monolayers to bundled nanomesh structures.[36] This process also simultaneously enhanced electrical conductivity and optical transparency. Furthermore, plasma-treated self-assembled gold nanowire membranes could be transferred to porous alumina oxide substrates and then elastomeric PDMS surfaces in order to build transparent and stretchable EDLCs.[37] Besides, stretchable supercapacitor electrode composed of silver nanowire (AgNWs) networks embedding in polyurethane acrylate (PUA) matrix was also reported.[38] The AgNWs were electro-coated separately with nickel and iron as positive electrode and negative electrode to establish transparent Ag-Ni $NWs_{(+)}$//Ag-Fe $NWs_{(-)}$ asymmetric supercapacitor with extended potential range up to 1.7 V and 35% of stretchability. In addition, metal mesh was also used as stretchable current collector. Conductive stainless filter meshes woven from 50 μm wires by plain weaving technique could possess stretchability up to 40% and could be electro-deposited other active materials easily.[39]

As for the metal compounds that serve as electro-active materials in supercapacitor electrodes, numerous of which have been studied over the years including metal oxides, metal sulfides, metal hydroxides and so on.[40,41] Compared to conventional carbon-based materials, metal compounds have higher theoretical specific capacity originating from redox reactions during electrochemical process that provide high pseudo-capacitance. However, the use of metal compounds in stretchable energy device is limited, and not so many of them have been introduced in the stretchable supercapacitor system, because of the inherent stiffness and brittleness which will cause electrochemical performance degradation under mechanical deformation. Metal compound-based materials are constantly combined with carbon materials or conductive polymers that help to increase their conductivity and flexibility. For instance, MoS_2 (molybdenum disulfide) have been combined with CNT by dip-coating hydrothermal-synthesized MoS_2 nanosheets solution on CNT/PDMS composite film to build stretchable electrodes.[42] As shown in **Figure 4**a, the MoS_2 nanosheets with large surface area not only showed high double-layer charge storage, but could also delivery excellent additional pseudo-capacitance. The as-prepared supercapacitor can deliver stretchability of 240% and possessed volumetric specific capacitance of 13.16 $F\cdot cm^{-3}$. In another example, MoS_2 was *in-situ* grow between holes and surface located inside graphene foam from CVD (chemical vapor deposition) using nickel foam as templates (**Figure 4**b).[43] The as-obtained compact graphene/MoS_2 composite films were transferred to the elastic PDMS substrates to establish stretchable supercapacitors with stretchability of 100% and volumetric specific capacity of 19.44 $F\cdot cm^{-3}$. Besides, MnO_2 (manganese dioxide) is another popular metal compound that has also been commonly utilized in stretchable supercapacitors. Here we provide one example as shown in **Figure 4**c-e, CNT films electrodeposited with MnO_2 and Fe_2O_3 respectively were transferred to pre-strained PDMS substrates to build asymmetric stretchable supercapacitor that achieved 100% of stretchability with extended voltage range.[44]

2.3 Conductive polymers

Conductive polymers such as polyaniline (PANI), polypyrrole (PPy), polythiophene (PT), PEDOT (poly(3,4-ethylenedioxythiophene)) as well as their derivatives are another type of popular materials in the stretchable supercapacitor field in that they not only provide pseudo-capacitance due to the redox reaction but also help improve the conductivity of the electrodes. They can be used solely as electro-active materials or combined with carbon materials and metal oxides in order to improve their electrochemical performances. As aforementioned in **Figure 1**c, the electrodeposition of PANI on wavy CNT film could further increase the specific capacity. As shown in **Figure 5**a, researchers directly dip-coated conducting polymer poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) on polyurethane (PU) fibrous nonwoven. The as-obtained PEDOT:PSS@PU nonwoven had stretchability exceeding 200% which could serve as stretchable conductors with tunable conductivity.[45] In another report, pre-strained SIBS (styrene-*block*-isobutylene-*block*-styrene) substrate was sputtered with 30 nm-thick gold film and electrodeposited PPy to prepare stretchable electrodes with buckled structure (**Figure 5**b).[46] The as-prepared supercapacitor could retain 81% of the initial capacitance after being stretched with 30% of strain for 1000 cycles. In addition, PPy and MnO_2 were electrodeposited on CNT film respectively to build stretchable asymmetric supercapacitor, as shown in **Figure 5**c.[47] Moreover, PEDOT was deposited on the ultrathin SWCNT (single-walled carbon nanotube) film through cyclic voltammetry (CV) method (**Figure 5**d). The as-obtained SWCNT/PEDOT hybrid film was merely 150 nm thick which led to ultrathin epidermal supercapacitor.[48] Recently, an all-in-one fiber with high conductivity and flexibility was prepared through wet-spinning of sulfuric acid treated PEDOT:PSS. By wrapping fibers around stretchable substrates, the established linear supercapacitor achieved 500% of high stretchability.[49]

To date, carbon-based material, metal-based material and conductive polymers are the three main choices for constructing stretchable electrodes. Each type of materials has its own pros and cons. Metallic materials have high intrinsic conductivity, which is favorable for fast charge/discharge but usually has low capacitance achieved; carbon-based materials often have high surface areas leading to high capacitance but material processing and conductivity may be limitations; conductive polymers offer good mechanical flexibility but the conductivity is usually not ideal. It is perhaps simple to conclude that we can combine these materials for better stretchable supercapacitors, however, it is non-trivial to control materials interface to prevent materials delamination/crack under mechanical deformation including bending and stretching.

3. Designing Technique Considerations for Stretchable Supercapacitors

In addition to material aspects, the layout design is another key parameter to achieve highly stretchable supercapacitors. Generally speaking, stretchable supercapacitors could be categorized into three types: one dimensional (1D) linear supercapacitors, two-dimensional (2D) planar supercapacitors (also includes microsupercapacitors), and three-dimensional (3D) stereo supercapacitors. Each type requires the maintenance of high conductivity of electrodes under mechanical deformations, otherwise, charging/discharging performance will be significantly deteriorated. Most state-of-the-art strategies are developed to minimize the strain concentrating on the electro-active layers. Representative techniques such as fiber winding, prestraining-then-buckling and island-bridge with serpentine interconnection designs are developed to achieve stretchability without sacrificing much of the electrochemical performances.

3.1 1D stretchable supercapacitors

1D supercapacitors, which can be described as fiber, wire or yarn-shaped supercapacitors, have received tremendous attention in recent years as they possess almost all the merits of conventional planar supercapacitors. More important, they can be processed into almost any desired shape, knotted or weaved into textile as well as easily connected into parallel or series, which are particularly suitable for wearable electronics.[50] Many 1D supercapacitors have been reported to date, however, only a small fraction of them were stretchable. In general, stretchable 1D supercapacitors are usually fabricated based on fiber electrodes/supercapacitors intertwined around another stretchable polymer wire, or directly depositing electrode materials on the (pre-strained) stretchable fiber cores. According to all the stretchable 1D supercapacitors reported to date, there are roughly four types of stretchable supercapacitors based on the design topologies: parallel, twisted, coaxial and helix.

As shown in **Figure 6**a and b, CNT sheets were deposited parallel on the opposite sides of a 300% pre-strained rectangular cross-section Ecoflex fiber, which formed buckled structure after releasing the strain.[51] Such sandwiched fiber could be enormously twisted and stretched without electrical shorting or significant resistance increase and could be woven into gloves (**Figure 6**c). The as-obtained supercapacitors showed outstanding mechanical stability which remianed electrochemical performances under deformations including bending, twisting and stretching up to 200% under both static and dynamic conditions. In another example, CNT fiber was synthesized with an extra oxidized procedure so that the CNT fiber has better ion-accessibility and increased hydrophilicity surface.[52] In order to build stretchable supercapacitors, two CNT fibers deposited with MnO_2 nanoparticles were laid parallel on a pre-strained PDMS film and covered with LiCl-PVA gel electrolyte as shown in **Figure 6**d. The supercapacitor delivered 40% of stretchability and high specific volumetric capacitance of around 409.4 $F \cdot cm^{-3}$ (or 133 $mF \cdot cm^{-2}$) at 0.75 $A \cdot cm^{-3}$. Parallel fiber supercapacitors can also be achieved by wrapping PPy-electrodeposited CNT films into composite fibers and assembling them parallel on PDMS substrates. Detailed tests for mechanical and electrochemical properties under both static/dynamic states was also carried out.[53]

Twisting-based configuration has been widely adopted in many early designed 1D supercapacitors,[54-56] most of which are flexible but not stretchable. Recently, it was shown that overlapped CNT sheets could be wrapped around pre-strained elastic fibers (64% polyester and 36% polyurenthane) (**Figure 6**e) and coated with a thin layer of PEDOT-PSS to further enhance the conductivity and electrochemical performance.[57] Two as-obtained electroactive fibers were twisted together with PVA/H_3PO_4 hydrogel served as both electrolyte and separator, as shown in **Figure 6**f. The as-prepared supercapacitor could tolerate stain of up to 350% with almost no change in specific capacitance (about 30.7 $F \cdot g^{-1}$). In addition, researchers creatively developed ultra-stretchable electrodes based on microscopically buckled and macroscopically coiled designing strategies which include pre-strained method and twisted configurations.[58] Core silicone rubber fiber was first coiled up to about 4000 $turns \cdot m^{-1}$ and then fully stretched to 1000%, after which wrapped with CNT layer. As a result, microscopic buckles and macroscopic coils were simultaneously formed along the fiber during relaxation (**Figure 6**g), which render the as-fabricated electrodes with ultra-stretchability of up to 600% to 800%. Similar yarn stretchable supercapacitor with coiled structures was also reported which could be hand-woven into a textile.[59]

Coaxial is another popular and representative type for 1D linear supercapacitor. In order to build coaxial stretchable supercapacitor, elastic polymer fibers are commonly used as the beginning core substrates. As shown in **Figure 7**a, CNT sheets were wrapped around elastic core rubber fiber and coated with H_3PO_4/PVA gel electrolyte step by step to build stretchable coaxial 1D stretchable supercapacitor, with clear cross-section SEM image shown in **Figure 7**b.[60] The resulting supercapacitor could be stretched to 75% of tensile strain without obvious degradation in specific capacity (18 F/g) and remain good capacity retention under repeated stretching. In order to further enhance the performances of fiber supercapacitor, another improved design was later proposed. Pre-strained core fiber and PANI electrodeposition were added to the coaxial configuration with similar fabrication process (**Figure 7**c).[61] By optimizing the pre-strain, CNT sheet thickness and PANI electrodeposition time, the overall stretchability could reach as high as 400% with specific capacitance of 111.6 $F\cdot g^{-1}$ or area specific capacitance of 3.08 $mF\cdot cm^{-2}$ under both static and dynamic strains. In addition, the as-prepared coaxial fiber stretchable supercapacitors could be woven across each other into stretchable power textiles as shown in **Figure 7**d. Recently, a distinctive asymmetric fiber supercapacitor was developed without a stretchable polymer core. Instead, a CNT fiber twisted from CNT strip was decorated with MnO_2 nanoflower (CNT@MnO_2) and used as core positive electrode. With gel electrolyte in between, CNT@PPy film was wrapped outside as negative electrode, as shown in **Figure 7**e.[62] The stretchability was achieved simply by further twisting the coaxial fiber supercapacitor into helical structure (**Figure 7**f and g). Although the stretchability is limited with about 20% of strain, but the potential window had been greatly extended up to 1.5 V with improved areal specific capacitance of 60.43 $mF\cdot cm^{-2}$.

The last type of stretchable 1D supercapacitors is the helix or spring-like configuration which is usually based on wrapping fiber electrodes around stretchable polymer core. As mentioned before, many flexible fiber electrodes fabricated to date are not stretchable, wrapping them into helix or spring-like structure is a good way to render the intrinsically non-stretchable fiber supercapacitor with novel stretchability.[63] As shown in **Figure 7**h, it has recently been demonstrated that fiber supercapacitors with ultrastretchability could be obtained by combining both twisted and helix designs.[64] Firstly, hybrid CNT/graphene/PANI wires were synthesized and twisted to construct fiber supercapacitor. Then, the as-prepared fiber supercapacitor was coiled into spring-like configuration. The electrochemical performance was well-preserved under maximum strain of 800% with specific capacitance of 138 $F\cdot g^{-1}$ at current density of 1 $A\cdot g^{-1}$. **Figure 7**i shows another spring-like supercapacitor using PPy-decorated rGO/MWCNT composite fibers.[65] The spring-like fibers were coated with self-healing polymer which achieved both stretchability and self-healability. The as-prepared supercapacitor could be stretched up to 100% with 82.4% capacitance retention. Besides, the sealed stretchable supercapacitor could be further applied as a ring or a wrist band wearable power device or integrated into a stretchable textile.

In conclusion, 1D linear stretchable supercapacitors can be achieved using parallel, twisting, coaxial or helix configurations with assistance of methods such as pre-strain, self-twisting and helix winding. Remaining challenges include high risks of electrical shortening for longer fiber, complicated electrodes assembling procedures. Some encouraging progress has recently made in integrating fiber stretchable supercapacitors into textiles to power wearable sensors/LED displays.[61] Further efforts may be devoted to improved durability and simplified preparation of 1D stretchable supercapacitors so that their integration with textiles can be seamlessly realized in real world.

3.2 2D stretchable supercapacitor

2D planar supercapacitors dominate both in scientific research and practical applications. Developments including new materials synthesis or novel structural research in supercapacitor area are most likely involved in the planar configuration. In order to achieve stretchability, buckled/wavy-structured electrodes through pre-strained substrates or solid electrolytes remain as the first choice for planar supercapacitor, similar to the pre-strain method used in 1D supercapacitors. Besides, some intrinsically stretchable substrates such as textiles or elastomers deposited with carbon materials or nanostructured metal can also be utilized directly in planar stretchable supercapacitors.

As an example, vinyl hybrid silica nanoparticle reinforced hydrogel polyelectrolyte (VSNPs-PAM) was reported to achieve an unprecedented stretchability of around 1500%.[66] By applying the prestrain-then-buckled method with CNT/PPy composite film as electrodes, the as-prepared wavy-structured stretchable supercapacitor could be stretched to 1000% of strain with over two times of capacitance improvement due to the increased contact areas between electrodes and the electrolyte (**Figure 8**a and b). In another report, through prestrain-then-buckling process, uniformly pre-stretched silicon rubber substrate was attached with CNT film to form buckled structure, which achieved an omnidirectionally stretchable supercapacitor, as shown in **Figure 8**c.[67] The supercapacitor remained its electrochemical performance under elongations of 200% regardless of whether uniaxial, biaxial or omnidirectional strain with specific area capacity of 9.52 $mF \cdot cm^{-2}$ at scan rate of 50 $mV \cdot s^{-1}$. In addition, Ecoflex substrate was also biaxially stretched and transferred with CNT/PEDOT film, as shown in **Figure 8**d and e.[68] There are also many other stretchable planar supercapacitors based on similar design, making the prestain-then-buckling method most popular in preparing 2D stretchable supercapacitors.[69-71] On the other hand, new design was proposed to fabricate 2D auxetic stretchable rGO/CNT networks through directional crystallization, freeze drying and radial compression procedures (**Figure 8**f).[72] The as-synthesized rGO/CNT networks were porous with honeycomb-like structures and was mechanically engineered into transversely isotropic 2D auxetic cellular architecture to serve as omnidirectionally stretchable electrodes (**Figure 8**g, h). The as-build EDLC exhibited tolerance to biaxial strain of 100%.

There are also some other planar stretchable supercapacitor systems based on intrinsic stretchability of substrates or nanostructured carbon/metal materials. For example, textile-based substrates are flexible with large surface area. Due to the knitted structures, some textiles could possess certain degree of stretchability which can be directly utilized to build stretchable planar supercapacitors. As shown in **Figure 9**a, cotton textiles were dip-coated with CNT and decorated with MnO_2 nanoparticles to serve as stretchable electrodes, but only with limited stretchability (around 20%).[73] In another demonstration, pristine textile with weft knitted structure was controllably carbonized to produce conductive textile (**Figure 9**b), which showed high electrical conductivity under strain up to 70%. The as-obtained supercapacitor could sustain tensile strain of 50% with specific capacitance of 246.3 mF/cm^2 (41.6 F/g).[74] In addition, the combination of elastomers and carbon or metal nanomaterials also results in intrinsically stretchable electrodes. As mentioned before in **Figure 4**a (also **Figure 9**c), supercapacitor based on CNT and MoS_2 coated on PDMS substrates could achieve 240% of stretchability,[42] while the CVD-synthesized graphene foam combined with MoS_2 (**Figure 4**b, **Figure 9**d and e) on PDMS substrates realized 60% of stretchability.[43] In another report, laser-induced graphene (LIG) was produced on polyimide sheet and peeled off by PDMS to build stretchable electrode.[75] As for intrinsically stretchable metal nanomaterials, our group fabricated ultrathin gold nanowire-based conductive thin film through Langmuir-Blodgett technique and transferring self-assembled monolayer gold nanowires (AuNWs) on PDMS substrate (**Figure 9**g).[37] The bundling AuNWs could form uniform wrinkle structure under repeated stretching remained conductivity as shown in **Figure 9**f. Stretchable EDLCs were obtained by assembling two conductive film with electrolyte in between, with negligible changes in resistance under strain up to 30%.

3.3 Micro-supercapacitors (MSCs)

Micro-supercapacitors (MSCs) have emerged as a new branch of planar supercapacitor in recent years.[76] Comparing to conventional planar supercapacitors, micro-supercapacitors require no separators, reduce the size and thickness of the whole system and allow facile on-chip integration with other electronic units due to their 2D configurations.[77,78] For stretchable micro-supercapacitors, island-bridge layout with serpentine interconnection design have been widely adopted to reduce the impact of applied tension strain on electrochemical performances. In one demonstration, sputtered gold-based current collectors were processed into serpentine interconnections which could afford the micro-supercapacitors to be stretched to 30% (**Figure 10**a).[79] In another report, a hybrid approach was utilized which combined soft PDMS/Ecoflex substrates and stiff PDMS island arrays where PET (polyethylene terephthalate) patches with micro-supercapacitors could be attached, as shown in **Figure 10**b.[80] In this way, the strain applied to the system was accommodated by the soft thin film so that the influence on MSCs was minimized. The strain distribution calculated by FEM (finite element method) analysis in **Figure 10**c also verified that under 40% of uniaxial strain, the island where micro-supercapacitor locate only hold 7% of strain while the space between islands reached up to 120%. In another demonstration, stretchable Ecoflex was chosen as substrates with relatively stiff PET films implanted, which formed strain suppressed zone so that the MSCs could be exactly placed and isolated from the strains.[81] Besides, the liquid metal (Galinstan) and Ag nanowires were used to link individual MSCs. The as-obtained MSC arrays have extended stretchability of 100% under uniaxial strain and 50% under biaxial strain. Recently, 3D-printing approach was introduced to fabricate patterned mold that could produce wavy structured PDMS electrode. With sputtered gold as current collector and MWCNT/PANI composite slurry as active materials, the MSC arrays could achieve high areal specific capacitance of 44.13 $mF\cdot cm^{-2}$ and stretchability of 40% (**Figure 10**d).[82]

There are some other stretchable MSC configurations reported. As shown in **Figure 11**a, tripod-structured PDMS was developed as stretchable substrate.[83] The tripod-structured PDMS was first stretched and transferred with GO (graphene oxide) microribbons and then released. The as-obtained system provided suspended wavy structured electrodes that ensure the stability of the MSCs under stretching/relaxing processes while the gap between each tripod also provided free space to relieve tension strain (**Figure 11**b). In another micro-supercapacitor system, honeycomb-shaped PDMS substrates were fabricated, with intrinsic deformation mechanism that localized large strain at the corners and low strain in large regions, as shown in **Figure 11**c and d.[84] MSC arrays were loaded in low-strain area and suffer minimum influence from the stretching strain. The achieved stretchablity was 150% with no obvious electrochemical performance degradation (specific volumetric capacitance of 1.86 $F\cdot cm^{-3}$ for single MSC).

3.4 3D stretchable supercapacitor

In addition to conventional stretchable supercapacitor configurations introduced above, including 1D, 2D supercapacitors and micro-supercapacitors, there are distinctive stretchable supercapacitor systems that also achieved remarkable stretchability and even 3D stretchability through kirigami or patterning-based editable techniques. For example, kirigami-based editing process was conducted by cutting linear patterns on as-fabricated planar supercapacitor that could transform the supercapacitor sheet into a honeycomb-like structure under tensile stretching as shown in **Figure 12**a. The supercapacitors could be stretched under a maximum strain of 400% with superior electrochemical stability and achieve high areal capacitance of 230.5 $mF\cdot cm^{-2}$ at current density of 1.6 $mA\cdot cm^{-2}$.[85] Furthermore, other 2D or even 3D stretchable architectures could also be easily constructed from this kirigami-based editable strategy (**Figure 12**b). In another report, CNT films were synthesized by CVD (chemical vapor deposition) on silica wafers with cellular patterned catalyst.[86] With PANI electrodeposition, the capacity could reach 42.4 $F\cdot g^{-1}$ for gravimetric capacitance (or 72.9 $mF\cdot cm^{-2}$ for the areal capacitance) while the uniaxial stretchability could reach 140%. It was straightforward to interconnect the cellular patterning supercapacitors in parallel or series so that both voltage and current could be tuned. In practice, the cellular supercapacitor served as a “watch strap” and could drive the commercial electronic watch (**Figure 12**c). By similar patterning with pyramid structure, 3D stretchable supercapacitor was established as shown in **Figure 12**d.[87] In addition, origami-type supercapacitor was also developed based on isolated graphite electrodes and sectionalized gel electrolyte (**Figure 12**e).[88] Graphite-based isolated electrodes was first synthesized on a cellular paper substrate and then deposited patterned gel electrolyte on the back sides so that the isolated graphite electrodes and sectionalized gel electrolyte were combined. In this design, the origami-type supercapacitor could sustain deformation from -60% of compression to 30% of stretching.

The state-of-the-art design of 1D, 2D and 3D supercapacitors are described above. Although stretchability can be achieved in either of the designs, their practical applications may need to be better defined. 1D stretchable supercapacitors possess native advantages to be sewn into textiles and integrated into everyday clothes; 2D stretchable supercapacitors may be suitable for epidermal 'tattoo-like' powering devices; 3D stretchable supercapacitors may be not so suitable for wearable energy applications because of their bulkiness. In the literature, volumetric capacitance, specific capacitance, and areal capacitance have been used in specific publications. Each one compares the performances based on specific circumstances. In our viewpoint, areal capacitance should be compared for wearable applications since large volume and weight are not favored; volumetric and specific capacitance may be key parameter for applications in electric cars but not important for wearable/implantable electronics.

4. Multifunctional Stretchable Supercapacitors

Highly stretchable supercapacitors offer great possibilities of versatile implications in the next-generation portable/wearable/implantable energy devices. For those applications, a number of other features are also essential, such as lightweight, ultrathin, conformable, transparent, self-healable and responsive.[89] In recent years, researchers have passionately attempted to incorporated such multi-functionalities into stretchable supercapacitor system.[90,91]

Ultrathinness is one essential parameter for stretchable supercapacitors, which meet the requirement for supercapacitors to be easily attachable on human skin conformably. In the aforementioned literature as shown in **Figure 13**a, researchers utilized ultrathin CNT film deposited with conductive PEDOT as electrodes to build supercapacitor system with only 1 μm of thickness. The as-prepared supercapacitor could be seamlessly attached to human skin as epidermal supercapacitor, while remained the configuration and electrochemical performance under deforming situations.[48]

Transparency is another attractive feature that has been widely introduced in many recently reported elctronic electrodes for flexible displays and windows, as a replacement of conventional rigid ITO-based electrodes.[34] In addition, transparent stretchable supercapacitors can realize invisible wearable electronics systems in practical applications. As mentioned in our previous chapters regarding to materials and configurations, many stretchable supercapacitors have also achieved transparency simultaneously in addition to stretchability, including both carbon and metal materials-based stretchable supercapacitors. As shown in **Figure 13**b, ultrathin graphene sheets through lamination provided the as-prepared stretchable supercapacitors with excellent optical transparency and mechanical stretchability.[32,33] As for metal-based materials, the aforementioned ultrathin gold nanowire-based stretchable supercapacitor we built was also transparent due to the monolayer assembly of ultrathin gold nanowires, as shown in **Figure 13**c.[37] Other metal nanowires including silver nanowires or gold-coated silver nanowire networks have also been constructed into transparent networks that serve as transparent stretchable electrodes with fine optical transmittance and mechanical stretchability (**Figure 13**d).[38,92] There are more examples of transparent stretchable supercapacitors based on both metal and carbon nanomaterials in the reported literatures.[93-95]

Self-healability as a remarkable function has been incorporated into stretchable supercapacitor system in recent years. In the previous example shown in **Figure 7**i, the as-prepared spring-like fiber stretchable supercapacitor was wrapped by self-healing polymer (PU, carboxylated polyurethane) as outer shell and could maintain 54.2% of its original capacitance after the third healing.[65] Furthermore, the aforementioned super-stretchable vinyl hybrid silica nanoparticle reinforced hydrogel polyelectrolyte had better self-healability through hydrogen bonds crosslinking, which could also utilized to build self-healable stretchable supercapacitors. The as-obtained supercapacitor maintained its electrochemical performances completely after 20 breaking/healing cycles and had excellent stretchability of 600%.[96]

5. Device-level Integration for Stretchable Supercapacitors

Towards real-world applications in wearable and/or implantable electronics, it is of vital importance to develop device-level integration technologies that incorporate stretchable supercapacitors with other electronic units to realize fully integrated, sustainable system.[97] It is encouraging to witness the progress made lately in developing cutting-edge integration technologies.[98-100]

Researchers have successfully combined sensors with energy harvesters to form the integrated electronics,[101,102] some of which contained stretchable supercapacitors as energy storage unit. As shown in **Figure 14**a, flexible yarn-shaped supercapacitors were fabricated using carbon fibers coated with PEDOT:PSS.[103] The as-obtained yarn supercapacitors were not stretchable intrinsically, but became highly stretchable up to 100% after being woven into the triboelectric nanogenerator (TENG) fabric through weft-knitting technique. In this case, the energy harvesters and the energy-storing supercapacitor were combined together, constituting the integrated stretchable fabric (**Figure 14**b). In another demonstration, the kirigami-based editable cutting technique was introduced to prepare the stretchable planar supercapacitors

(**Figure 14**c), with specific capacitance of ∼12 F/g and ∼1 mF/cm^2 at scan rate of 10 mV/s. The stretchable supercapacitor was further integrated with silicone rubber-based TENG with layer-by-layer framework to build the all-in-one shape-adaptive self-charging package of wearable/portable electronics (**Figure 14**d). This integrated system could harvest and convert mechanical energy from all kinds of human motions including bending, twisting and stretching into electricity and then store into stretchable supercapacitor unit, which showed promising improvements for future wearable integrated electronics.[104] Despite of encouraging progress made recently in developing integration technology for stretchable supercapacitors, it is still in the embryonic stage. Most of reports to date focused on the integration designs, preparation methods, and device flexibility, rather than quantitative performance evaluations under static/dynamic stretched states. This may be due to unsatisfactory stability and/or durability originating from materials delamination and/or cracks occurred at the soft/hard interfaces of multilayer materials and different layers of electronic units.

6. Summary and Perspectives

Stretchable supercapacitors could become indispensable components in future soft electronics system, which could also have the potential to transform the current bio-diagnostics system. In this review, we thoroughly discuss materials aspects in stretchable electrodes from carbon nanomaterials, metallic nanomaterials to conductive polymers. Each type of materials has respective pros and cons. While carbon-based materials (especially CNT and graphene) dominate in stretchable supercapacitors, metallic and polymeric materials have also been successfully used as current collectors or electro-active materials. Further improvements should be focused on conductivity enhancement, mechanical stability under practical dynamic deformations as well as rational combination of different materials. It is noteworthy that electrolyte is also an essential part in building stretchable supercapacitors. Most electrolytes used at present are PVA (polyvinyl alcohol)-based gel electrolyte with less stretchability and limited voltage window. Although some solid polyelectrolytes with impressive stretchability have been developed but with reduced voltage range,[66,96] demonstrating the challenges still remain.[105] Besides, the synergistic effects between electrode materials and redox electrolytes have rarely been studied in stretchable supercapacitor system.[106] In the future developments, novel electrolytes with ideal stretchability, electrochemical stability and extended voltage window may need to be paid more attention.

We also describe various designing strategies and techniques based on different dimensions of stretchable supercapacitors, classified into 1D linear, 2D planar and 3D stereo configurations. In particular, viable techniques including winding/twisting or coxial methods for 1D supercapacitor, prestrain-then-buckling method for both 1D and 2D supercapacitor, island-bridge with serpentine interconnection design for microsupercapacitor and kirigami/patterning-based editable method for 3D supercapacitor are described. Despite of encouraging progress made in the field of stretchable supercapacitors to date, it remains challenging to integrate them into day-to-day wearable, on-skin and/or implantable electronics system, since majority of current stretchable supercapacitors are still bulky especially for those 3D systems. Main efforts have been focused on stretchability and electrochemical performances rather than practical considerations. Future efforts should concentrate on integration with clothes, attachability to human skin or implanted in inner organs, and evaluate their performances in real-time and *in-situ*. Multi-functionalities such as ultrathinness, transparency and self-healability may also be additional must-have features in the future, bringing a step closer for biomedical energy solution to be true part of human body. Besides, soft/hard materials interface, device/human interface, and interconnect issues need to be paid more attention to prevent cracking, delamination or short circuit, which are the limiting factors for long-term stability and durability. For realizing real-world applications, multidisciplinary collaborations beyond materials scientists, nanotechnologiests and electronic engineers become increasingly more important.

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Conflict of Interest

The authors declare no conflict of interest.

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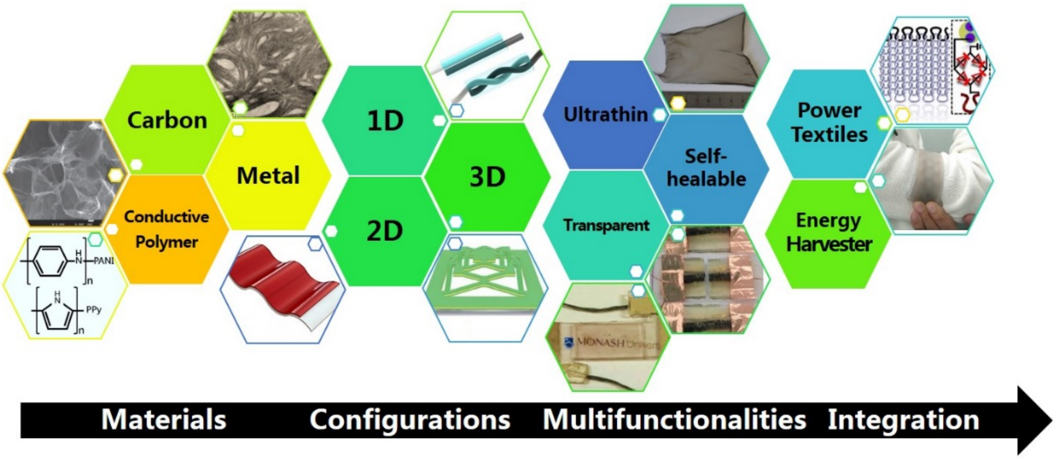
Review Figures



Scheme 1. Research on stretchable supercapacitor based on materials (carbon, metal and conductive polymer), configurations (1D linear, 2D planar and 3D stereo), multifunctionalities (ultrathinness, transparency and self-healability) and integration (power textiles and energy harvester). "carbon materials (3D graphene nanosheets)" reproduced with permission.[34] Copyright 2013, Wiley-VCH. "metal materials (ultrathin gold nanowire bundles)" reproduced with permission.[36] Copyright 2016, Wiley-VCH. 3D configuration "three-dimensionally stretchable supercapacitor" reproduced with permission.[87] Copyright 2016, The Royal Society of Chemistry. "Ultrathin functionality (ultrathin carbon nanotube film)" reproduced with permission.[48] Copyright 2016, Wiley-VCH. "Transparent functionality (transparent supercapacitor)" reproduced with permission.[37] Copyright 2012, Wiley-VCH. "Self-healable functionality (self-healable supercapacitor)" reproduced with permission.[65] Copyright 2017, American Chemical Society. "Power textiles (yarn-shaped supercapacitor sewn into textiles)" reproduced with permission.[103] Copyright 2017, American Chemical Society. "Energy harvester (planar supercapacitor combined with triboelectric nanogenerator)" reproduced with permission.[104] Copyright 2016, American Chemical Society.

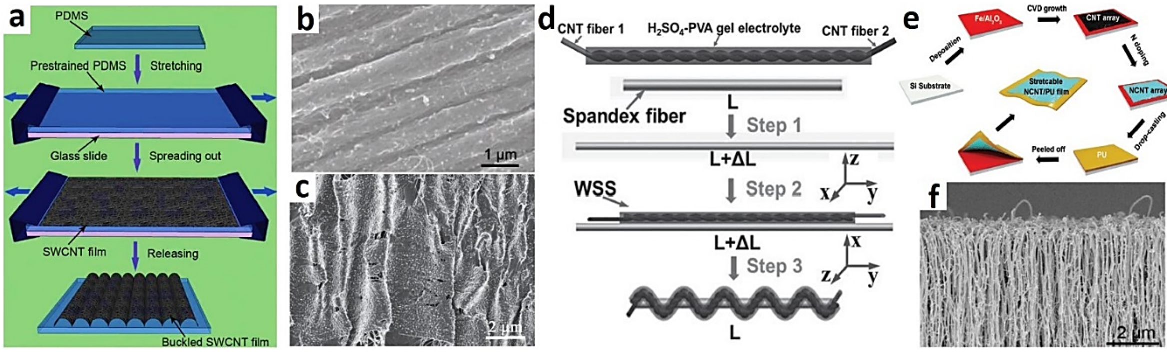


Figure 1. (a) Schematic illustration of preparation procedure for buckled SWCNT film on PDMS. (b) SEM image of buckled SWCNT film. Reproduced with permission.[28] Copyright 2013, Wiley-VCH. (c) SEM image of crumbled CNT/PANI composite. Reproduced with permission.[29] Copyright 2014, Wiley-VCH. (d) Schematics of the fabrication procedures for stretchable wire shaped supercapacitor. Reproduced with permission.[30] Copyright 2014, Wiley-VCH. (e) Schematic illustration to the preparation of elastic N-doped CNT/PU film. (f) SEM image of NCNT (20 min of re-growing). Reproduced with permission.[31] Copyright 2017, Wiley-VCH.

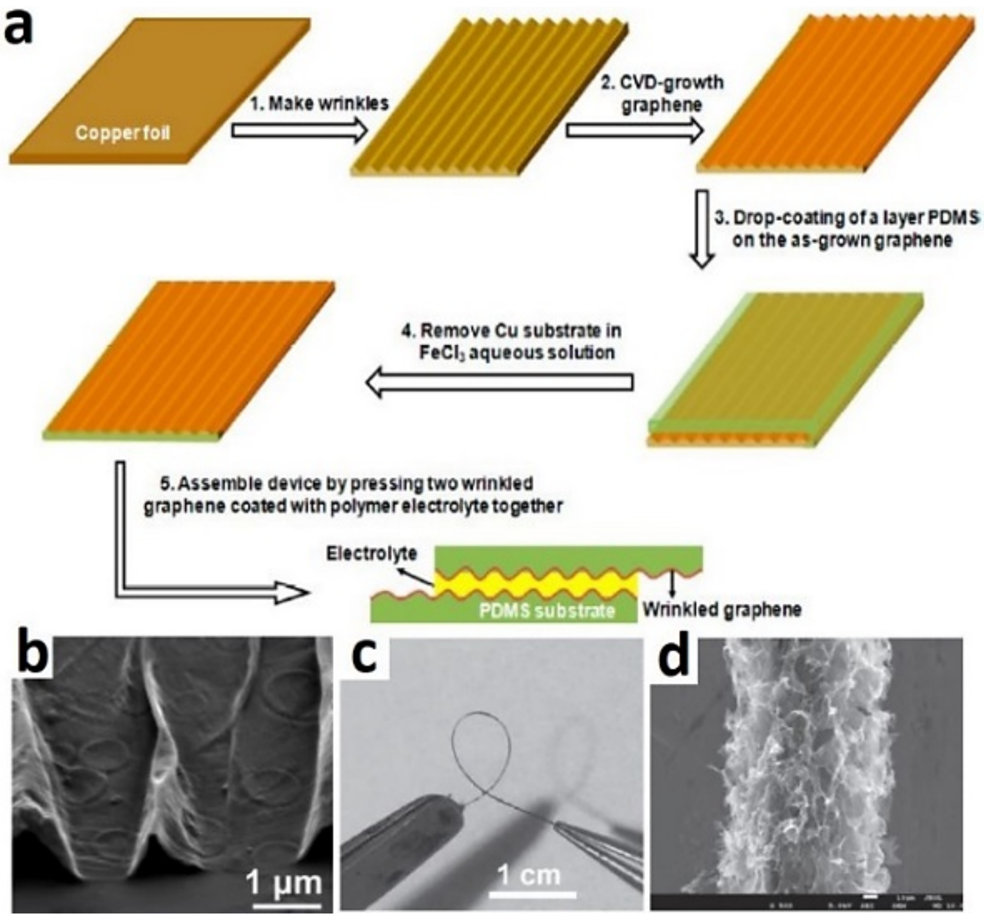


Figure 2. (a) Schematic illustration to fabrication procedures for transparent wrinkled graphene sheets and stretchable supercapacitors. Reproduced with permission.[32] Copyright 2013, American Chemical Society. (b) SEM image of buckled graphene film on PDMS substrate. Reproduced with permission.[33] Copyright 2014, American Chemical Society. (c) A photograph of GF@3D-G. (d) SEM image of GF@3D-G. Reproduced with permission.[34] Copyright 2013, Wiley-VCH.

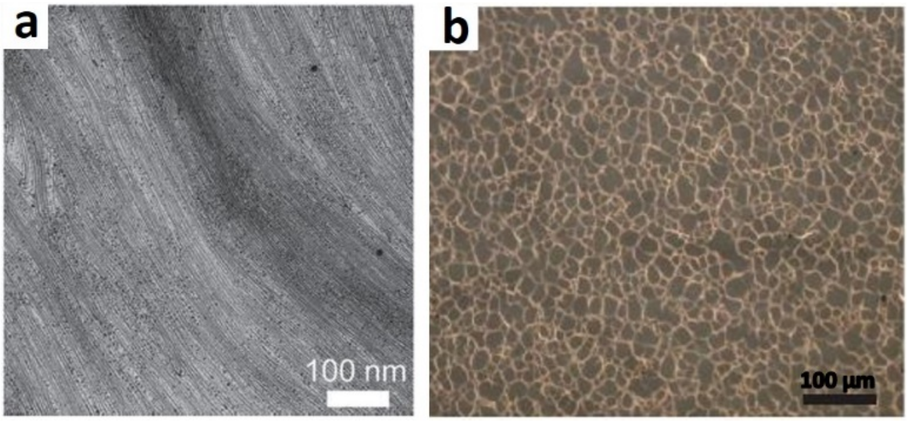


Figure 3. (a) TEM images of monolayer superlattice nanomembrane of ultrathin gold nanowires. Reproduced with permission.[36] Copyright 2012, Wiley-VCH. (b) Optical microscopy image of the gold nanowire mesh film. Reproduced with permission.[35] Copyright 2016, Wiley-VCH.

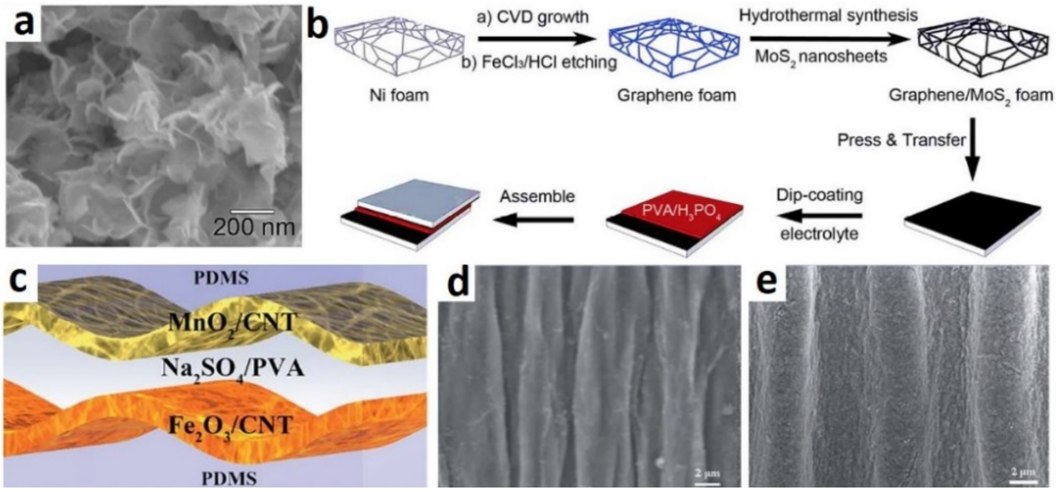


Figure 4. (a) SEM image of MoS_2 nanosheets. Reproduced with permission.[42] Copyright 2016, Wiley-VCH. (b) Schematic illustration of the fabrication of stretchable all-solid-state supercapacitor based on graphene/MoS_2 composite films. Reproduced with permission.[43] Copyright 2017, The Royal Society of Chemistry. (c) Schematic illustration of asymmetric stretchable supercapacitor based on wrinkled positive MnO_2/CNT hybrid film electrode and negative Fe_2O_3/CNT hybrid film electrode. (d,e) SEM images of wrinkled MnO_2/CNT and Fe_2O_3/CNT hybrid film. Reproduced with permission.[44] Copyright 2016, The Royal Society of Chemistry.

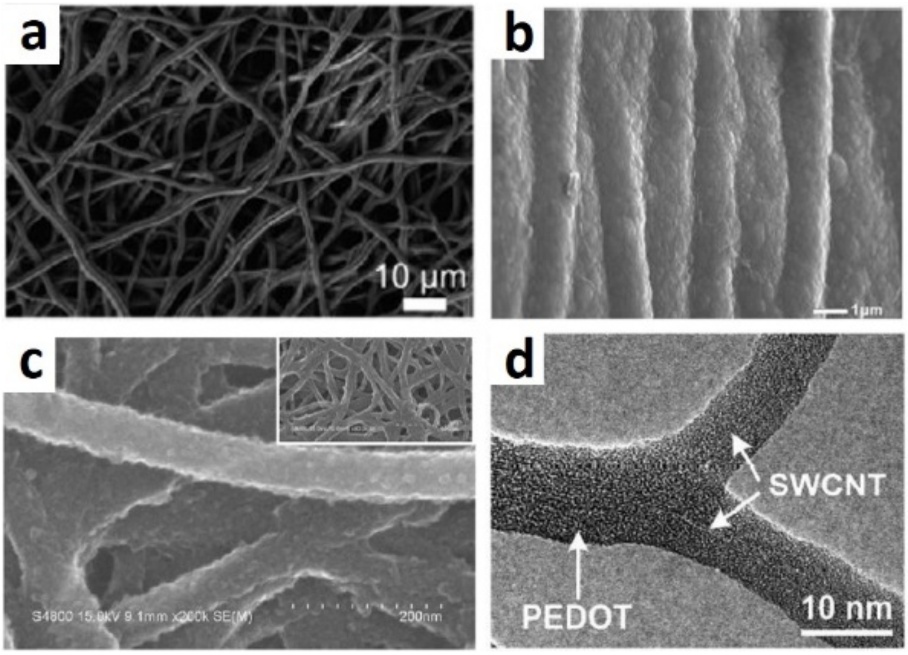


Figure 5. (a) SEM image of surface of PEDOT:PSS@PU film. Reproduced with permission.[45] Copyright 2017, American Chemical Society. (b) SEM image of buckled PPy electrode. Reproduced with permission.[46] Copyright 2013, American Chemical Society. (c) FE-SEM image of CNT@PPy film (inserted with the corresponding FE-SEM image of smaller magnification). Reproduced with permission.[47] Copyright 2015, American Chemical Society. (d) SWCNT (single-walled carbon nanotube) bundles deposited with PEDOT. Reproduced with permission.[48] Copyright 2016, Wiley-VCH.

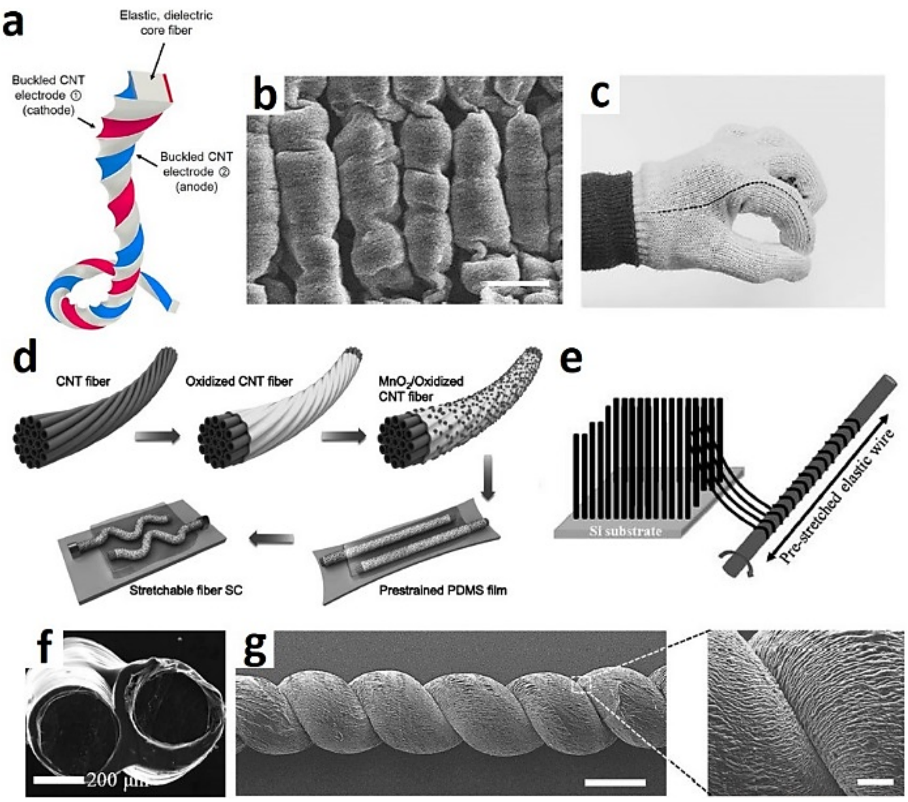


Figure 6. (a) Schematic illustration of twisted rectangular Ecoflex fiber core sandwiched by two symmetric buckled CNT electrodes. (b) SEM images of CNT buckles during fiber relaxation from the fabrication strain (scale bar 20 μm). (c) Stretchable sandwiched fiber woven into a commercial glove. Reproduced with permission.[51] Copyright 2016, American Chemical Society. (d) Schematic illustration of fabrication process of the stretchable fiber supercapacitor. Reproduced with permission.[52] Copyright 2017, Wiley-VCH. (e) Schematic illustration of wrapping aligned CNT sheet around a pre-stretched elastic wire. (f) SEM image of cross-section of the twisted wire-shaped supercapacitor. Reproduced with permission.[57] Copyright 2015, Wiley-VCH. (g) SEM images of coiled/buckled CNT electrode (scale bar 500 μm) and its magnification. Reproduced with permission.[58] Copyright 2017, Wiley-VCH.

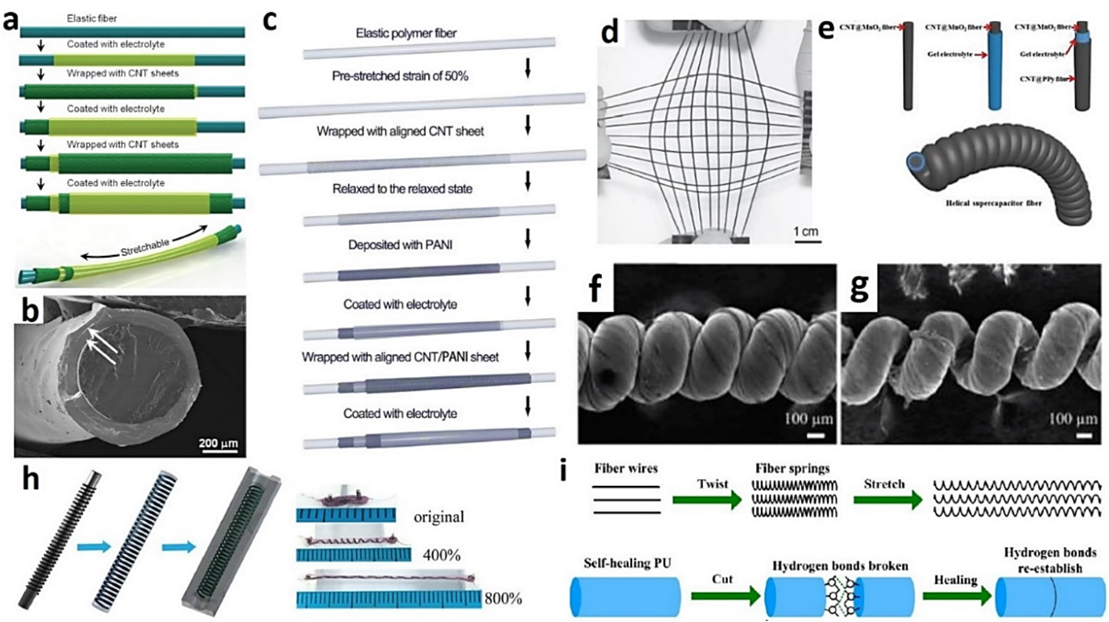


Figure 7. (a) Schematic illustration of fabrication of highly coaxial stretchable fiber-shaped coaxial supercapacitor. (b) Cross-sectional SEM image of the coaxial supercapacitor. The arrows show the aligned CNT sheet. Reproduced with permission.[60] Copyright 2013, Wiley-VCH. (c) Schematic illustration of the fabrication of fiber-shaped supercapacitor with prestrain design. (d) Stretchable textile consists of coaxial fiber supercapacitors under stretching by 100%. Reproduced with permission.[61] Copyright 2014, Wiley-VCH. (e) Schematic of helical coaxial fiber supercapacitor fabrication process. (f,g) SEM images of the stretchable asymmetric fiber supercapacitor under 0% and 50% strain. Reproduced with permission.[62] Copyright 2017, Wiley-VCH. (h) Schematic illustration of the fabrication of superelastic fiber-based supercapacitor and under different strain up to 800%. Reproduced with permission.[64] Copyright 2017, Wiley-VCH. (i) schematic diagrams of the manufacturing process and mechanism of stretchable and self-healable PPy/RGO/MWCNT electrodes. Reproduced with permission.[65] Copyright 2017, American Chemical Society.

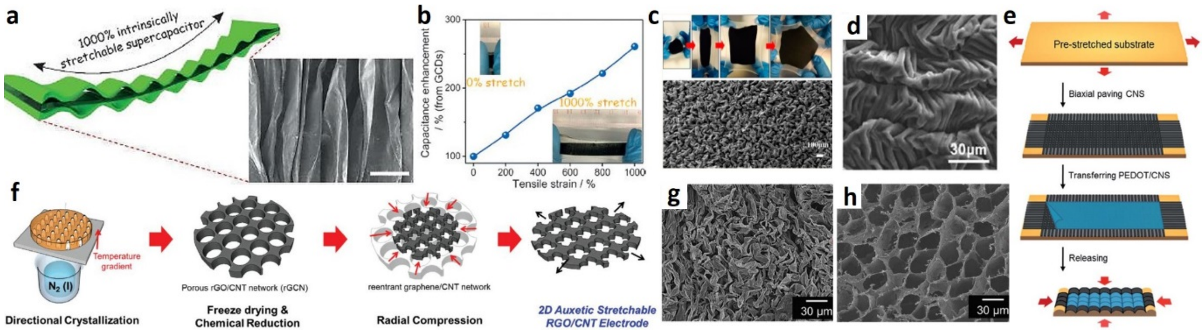


Figure 8. (a) Schematic of planar supercapacitor with 1000% of ultra-stretchablility and related SEM image of the wavy PPy@CNT paper electrode. (b) Capacitance enhancement under increase of tensile strain (insets: photographs of the supercapacitor at fully released state and under 1000% strain). Reproduced with permission.[66] Copyright 2017, Wiley-VCH. (c) Omnidirectionally stretchable supercapacitor under various stretching states and SEM image for buckled CNT film. Reproduced with permission.[67] Copyright 2016, American Chemical Society. (d) SEM image for biaxial wrinkled structure of the PEDOT/CNS hybrid nanomembrane. (e) Schematic diagram of the fabrication process of a stretchable PEDOT/CNS hybrid nanomembrane. Reproduced with permission.[68] Copyright 2016, The Royal Society of Chemistry. (f) Schematic illustration of the fabrication for omnidirectionally stretchable 2D auxetic reentrant graphene/CNT networks, with SEM micrographs of the networks after the directional freezing process (g) and after freezing process (h). Reproduced with permission.[72] Copyright 2017, The Royal Society of Chemistry.

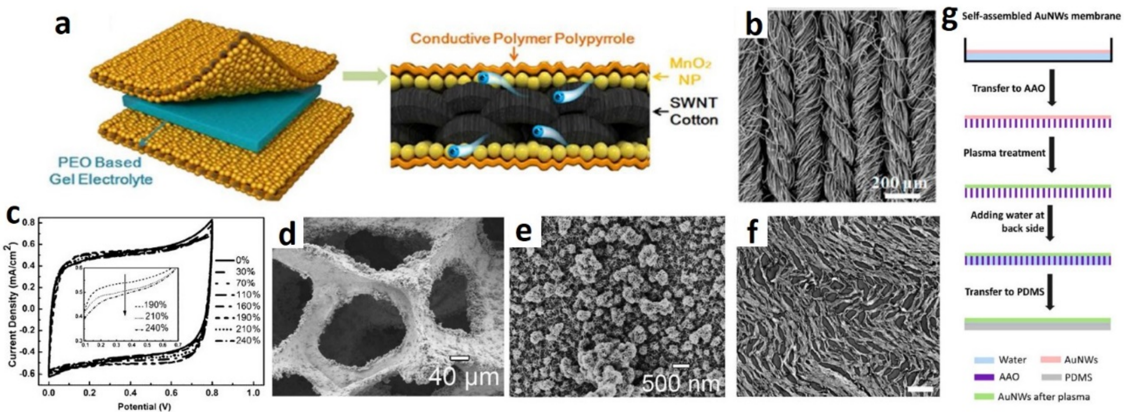


Figure 9. (a) Schematic illustration of the fabrication of PPy-MnO_2-coated textile supercapacitor. Reproduced with permission.[73] Copyright 2015, American Chemical Society. (b) SEM image of the carbonized textile. Reproduced with permission.[74] Copyright 2017, American Chemical Society. (c) CV curves of supercapacitor under different degrees of stretching varying from 0% to 240% at the scan rate of 0.1 V/s. Reproduced with permission.[42] Copyright 2016, Wiley-VCH. (d,e) SEM images of graphene/MoS_2 foams under different magnification (68.3 wt% of MoS_2). Reproduced with permission.[43] Copyright 2017, The Royal Society of Chemistry. (f) Wrinkle structure of AuNW film (Scale bar 500 nm) after repeated stretching process. (g) Fabrication process of stretchable and conductive self-assembled AuNWs electrode. Reproduced with permission.[37] Copyright 2016, Wiley-VCH.

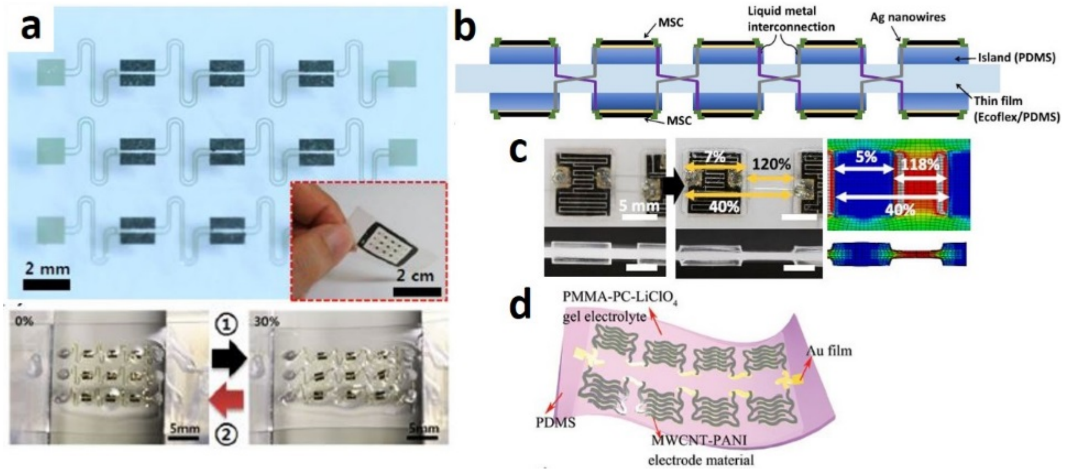


Figure 10. (a) Photographs of 2D micro-supercapacitor arrays on PDMS substrate with serpentine connection/island-bridge designs and stretched under 30% strain. Reproduced with permission.[79] Copyright 2013, American Chemical Society. (b) Schematic illustration for side view of integrated MSCs on stretchable PDMS/Ecoflex substrate. (c) Photographs of the stretchable MSC arrays under strain of 0% (left) and 40% (middle) and the corresponding strain distribution (right) based on FEM (finite element method) analysis. Reproduced with permission.[80] Copyright 2014, American Chemical Society. (d) Schematic diagram of the fabricated MSC arrays based on wavy structured PDMS. Reproduced with permission.[82] Copyright 2017, Wiley-VCH.

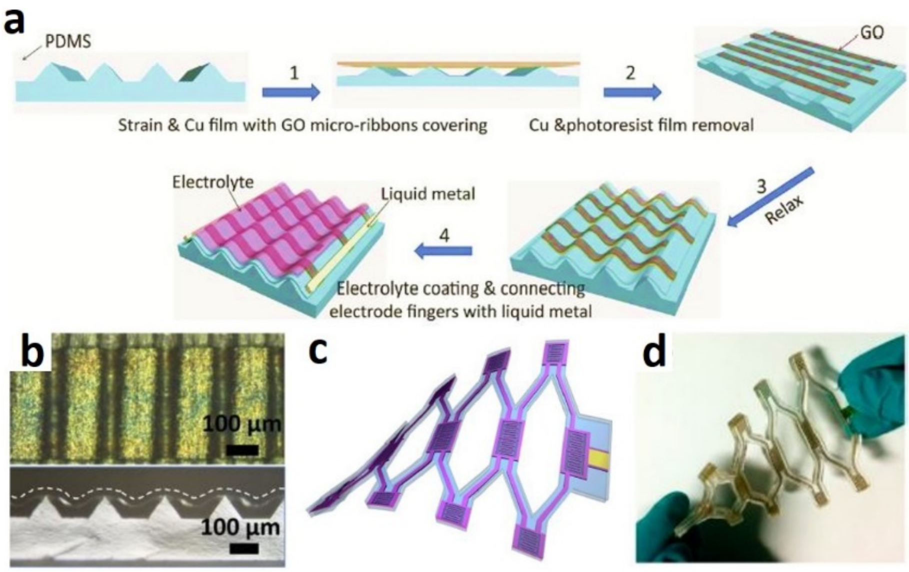


Figure 11. (a) Schematic illustration of transferring electrode arrays onto the tripod-structured PDMS substrate to stretchable micro-supercapacitors. (b) Optical image of as-obtained MSC (upper) and corresponding cross-section SEM image (lower). Reproduced with permission.[83] Copyright 2015, Wiley-VCH. (c) Schematic illustration of honeycomb structured MSC arrays under stretched and bent states. (d) Photograph of as-fabricated MSC arrays under stretched and bent states. Reproduced with permission.[84] Copyright 2016, American Chemical Society.

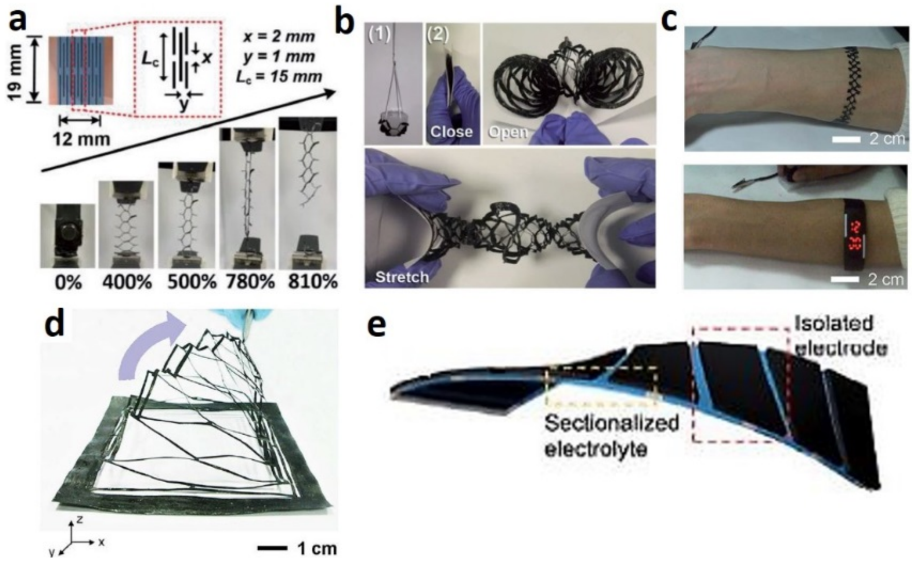


Figure 12. (a) Digital images of honeycomb-like supercapacitors through linear cutting strategy under different strain tests. (b) Editable supercapacitors with 3D basket structures under different states. Reproduced with permission.[85] Copyright 2017, Wiley-VCH. (c) "Watch strap" supercapacitor powering commercial watch. Reproduced with permission.[86] Copyright 2016, The Royal Society of Chemistry. (d) Photograph of pyramid-shaped CNT film under the simultaneous stretching along the x and z axes. Reproduced with permission.[87] Copyright 2016, The Royal Society of Chemistry. (e) Schematic for origami-type supercapacitor with periodically isolated electrodes (IEs) and sectionalized electrolytes. Reproduced with permission.[88] Copyright 2014, The Royal Society of Chemistry.

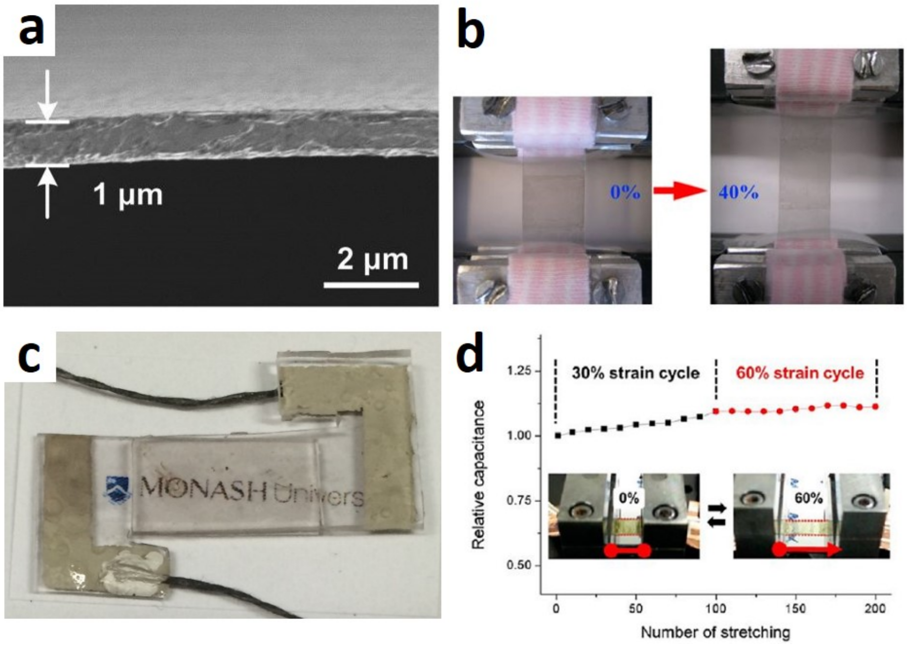


Figure 13. (a) Ultrathin supercapacitor configuration with 1 μm thickness. Reproduced with permission.[48] Copyright 2016, Wiley-VCH. (b) Digital photo of the graphene-based transparent supercapacitor stretched from the tensile strain of 0% to 40%. Reproduced with permission.[33] Copyright 2014, American Chemical Society. (c) Monolayer ultrathin gold nanowire-based transparent supercapacitor. Reproduced with permission.[37] Copyright 2016, Wiley-VCH. (d) Gold-coated silver nanowire-based stretchable transparent supercapacitor under 30% and 60% stretching and corresponding capacitance retention. Reproduced with permission.[92] Copyright 2016, American Chemical Society.

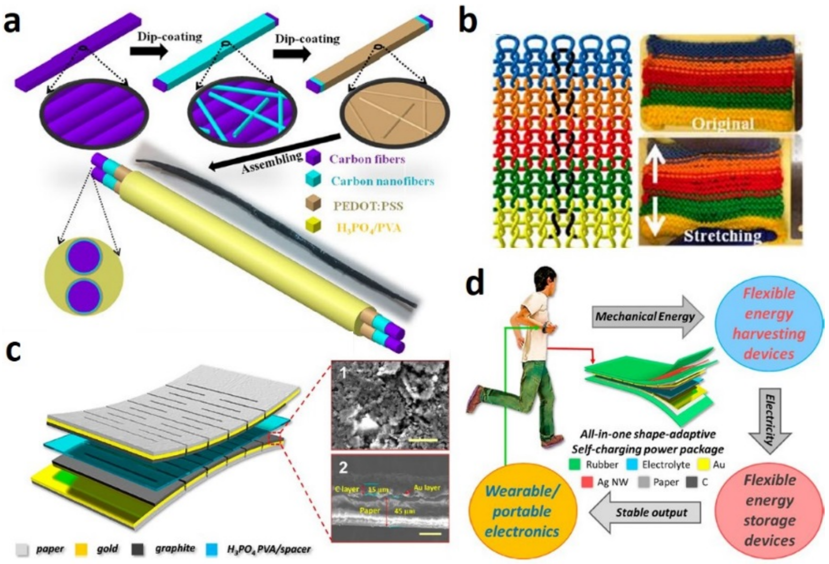


Figure 14. (a) Schematic illustration of yarn supercapacitor composed of two PEDOT:PSS/CNF/CF electrodes in parallel. (b) Yarn supercapacitors knitted into the normal knitting fabric at initial state (upper right) and stretched state (lower right). Reproduced with permission.[103] Copyright 2017, American Chemical Society. (c) Schematic structure of the kirigami-based supercapacitor. (d) Working mechanism of an all-in-one shape-adaptive self-charging package for wearable/portable electronics. Reproduced with permission.[104] Copyright 2016, American Chemical Society.

This review summarizes recent developments in stretchable supercapacitors for applications in future wearable electronics.