

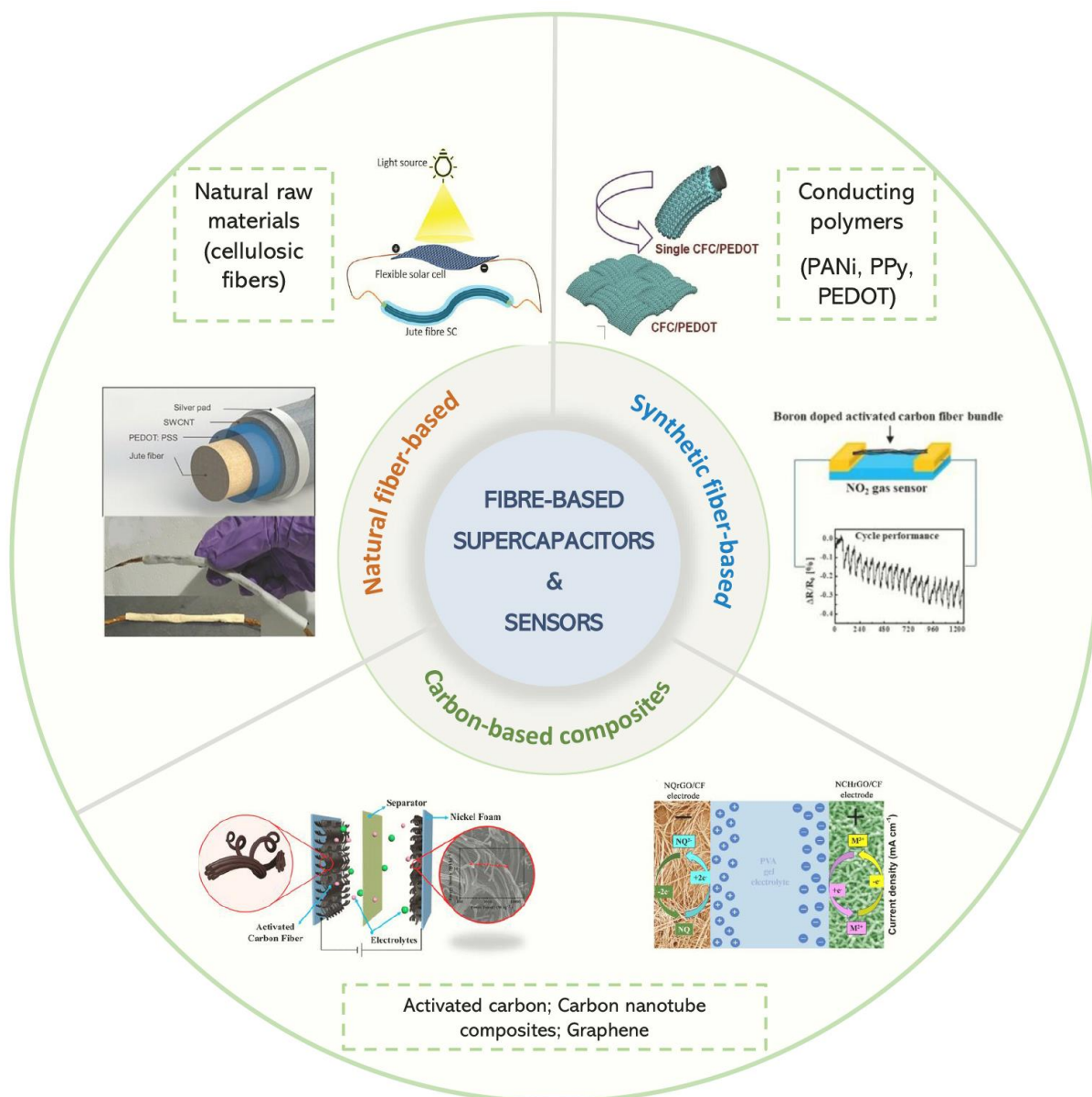
## **A review of Fiber-based Supercapacitors and sensors for energy-autonomous systems**

**Abstract:** Advancements in wearable technologies in the past few years have influenced the fabrication of fiber-based supercapacitors and sensors for next-generation energy-autonomous systems. There has been an increase in research on conductive electrodes on fibers for sensing ability and storing of energy based on their ease of fabrication, stretch and flexible abilities. These replace the conventional rigid devices that limit the flow of air and provide discomfort when used in clothing. Fiber-based sensors are generally fabricated to detect environmental and physiological changes in real-time. They are mostly fabricated for use as temperature, photo, chemical, and tactile sensors using different applications, and materials. A variety of wearable physical, chemical, biological, and optical sensors that have been described as self-powered or energy-autonomous in recent years rely on these technologies, as well as energy generators, electrochemical energy storage (EES) systems, wireless power technologies, self-powered sensors, and hybrid energy systems that combine energy generators and electrochemical energy storage. This paper highlights a comprehensive review of the recent advancements in fiber-based and sensors for supercapacitors energy-autonomous systems. The paper further highlights fiber-based material properties, such as lightweight, flexibility, high surface area, and their impact on energy density and stability. The paper also discusses the various fabrication methods for fiber-based supercapacitors and sensors, including electrospinning, dip-coating, and self-assembly. Finally, the paper elucidates future directions of fiber-based sensors and supercapacitors for electrochemical energy storage and visa vis sustainable production.

**Keywords:** Fiber based sensors, fiber-based supercapacitors; energy autonomous systems; wearable technologies; electrochemical energy storage.

## Graphical Abstract:

Fibers and their related composites have been considered for the fabrication of SCs and sensors, which find applications in energy-autonomous systems. Herein, we describe the fabrication methods and present recent advances in natural-based and synthetic-based composites used for SCs and sensors. Lastly, we highlight future research directions toward a more sustainable material and production.



## 1. Introduction

The rapid increase in population growth and its associated pollution of flora and fauna has given rise to global warming and an energy crisis resulting in health glitches over the past decades, hence recent developments in embedded active health monitoring devices into clothing to limit threats to sustainable societies. This has necessitated the design of smart embedded devices to sense the environmental and physiological body state [1]. For this, the design of an effective, yet, cleaner energy storage system is imperative [2, 3]. It is thus important that energy storage systems aid in solving intermittent power supply problems due to their reliable, stable, and highly efficient properties [4]. As such the emergence of supercapacitors (SCs) provided a potential energy storage technology over conventional batteries for their safe operations, low cost, easy fabrication, fast charging, and long cycle life [4]. SC are typically grouped as pseudo capacitors (fast reversible faradaic charge transfer on the electrode surface), electrochemical double layer capacitors (at the interface of the electrode and electrolyte surface with physical adsorption of ions) based on charge storage mechanism [5, 6] and hybrid capacitors (which combines the properties of the other two types) [7]. Electrode materials for SCs are grouped into electrochemical double-layer capacitors (EDLC) using carbon nanotubes, graphene and activated carbons; pseudo-SCs using conducting polymer and metal oxides; and hybrid SCs using asymmetric pseudo/EDLC, composite and rechargeable battery-type, based on how the charges are stored [8-10]. Alternatively, sensors are devices that effectively detect, respond, measure, and record inputs from the immediate physical surroundings. Typically, conventional sensors are rigid and hence limit their application in active monitoring in certain fields. Supercapacitors, often referred to as electrochemical capacitors or ultracapacitors, are one of the possible choices among energy-storage systems due to their many uses in portable gadgets, electric cars, and fixed energy storage systems. SCs offer three primary benefits; high specific power of around  $10 \text{ kW kg}^{-1}$ , extended cycle life  $>10^5$ , and quick charge/discharge operations occurring in only a few seconds [11, 12]. These benefits outweigh batteries and traditional capacitors for energy storage technologies.

In recent years, advances have been made in the fabrication of flexible, stretchable, and mechanically tough SCs for various applications such as energy autonomous systems (EAS). Typically, two-dimensional or basic planar configurations have been fabricated for flexible SCs using different electrode substrates such as conductive carbon fiber cloths and papers, and coated plastics [13, 14]. These fabricated flexible electrode substrates cannot be woven into fabrics, are uncomfortable to wear by humans, and blocks the flow of air when fixed on

cloth [15]. Intrinsically, fiber-based materials have seen great interest in the fabrication of SCs and sensors in recent years for wearable technologies [16, 17]. These are largely due to their breathability, stretchability, flexibility, comfortable handle, and relatively easy fabrication compared to their rigid convention counterparts. Furthermore, the possibility of fabricating these fibers into comfortable and flexible clothing through knitting or weaving has influenced their exposure to and exploration of wearable technology.

EAS is divided into three clear parts, thus, energy generation, energy conversion and optimization, and energy consumption. The EAS effectively harvests energy and converts it to power and operates low-consuming devices for effective feedback or response [18]. It is imperative to note that SCs store electrical energy efficiently and deliver quickly for use by devices that require power density while sensors convert chemical and physical stimuli into electrical signals for further analysis and processes by devices for EAS. The needed energy stored by the SCs is used to power the sensors to transmit the necessary signals for monitoring [19]. For EAS, both SCs and sensors are effectively integrated for appropriate energy delivery to power the sensing devices for reliable monitoring and feedback of activities in the environment. Alternatively, integrated sensors can be applied to measure the power stored and energy consumed to ensure the safety of the wearable system. Extreme heat and temperature produced from these SCs could pose some discomfort to wearers. These fiber-based sensors and SCs for EAS in wearable technology have increased their applications in critical areas. For instance, an eco-friendly EAS was developed using natural jute for humidity and temperature and humidity sensing [20]. Here, using the coating method, conductive materials are applied on the surfaces of the jute fibers. The fabricated fiber-based SCs when charged are used to power the fiber-based sensors for temperature and humidity sensing in the environment with great potential for wearables and environmental monitoring. Other recent advances have adopted varying fabrication methods including thermal drawing, deposition, and spinning in their manufacturing process [21].

Tremendous research efforts have been reported on the design, fabrication, and multi-functionalities of flexible fiber-shaped SCs [22], fiber materials for wearable electronics [23], fiber-based wearable SCs [13, 24], paper-based SCs based on cellulose [25], green biomass based composites for electrocatalysis, sensors and SCs [26], micro-SCs for alternating current filtering performance [27], energy systems and fiber-based sensors [21], wearable EAS sensors [28] and intelligent wearable sensors for rehabilitation purposes [29]. However, most of these reviews focus on specific fiber-based SCs or sensors for EAS with limited efforts to provide a comprehensive dichotomy of the two components of EAS in

wearable technologies. It is therefore imperative to provide a comprehensive review of the exigencies of SCs and sensors for fiber-based EAS. In this regard, this study presents a review of recent progress in fiber-based sensors and SCs that are applied in EAS. The review highlights the use of natural, synthetic, conducting polymer and their composite fiber-based materials for sensors and SCs for EAS, their methods of fabrication and also provides the future prospects and advancement in the field for integration in wearable electronics. Finally, future research development of fiber-based SCs and sensors for EAS are suggested to focus on key areas using chitosan-based or cellulose-based materials towards UN sustainable development goals.

## **2. Publication progress, top journals, and country collaborations**

The fabricated fiber-based sensors and SCs are effectively utilized for energy autonomous systems (EAS) which are capable of providing and storing information without any power source connected [18]. This has led to increased research output in the field of fiber-based sensors and SCs for EAS. A quantitative analysis as of July 24, 2023, using search blocks "fiber-based" AND "sensors" OR "supercapacitors" OR "energy-autonomous systems" in the Scopus database confirmed increased research output by leading researchers within the field of study. By limiting search results to journals (source type) and articles (document type), a resultant output of 2118 publications was obtained from the initial 3735 publications which comprised varying document types. In fact, the first publication as revealed from the search results was published in 1984 [30]. This study fabricated a gas sensor from optical fibers to remotely measure the absorption of low-level CH<sub>4</sub>gas within a near-infrared region.

Results from Figure 1a show a steep rise in research progress from the 2000s for flexible fiber-based sensors and SCs that can be easily integrated to form clothing for varying applications. Further analysis from Figure 1b shows the top contributing country was China, followed by the United States, India, the United Kingdom, and South Korea, all contributing above one hundred publications each. Subsequently, Figure 1c showed that the *IEEE Sensors journal* was the top leading publishing journal followed closely by the *Journal of Lightwave Technology* and *Optics Express* on flexible fiber-based sensors and SCs for EAS. This has largely influenced a bulk of collaboration between these countries as shown in Figure 1d to help advance material synthesis, improved performance, and usage of these fiber-based sensors and SCs for EAS. This collaboration analysis was conducted using Biblioshiny of the Bibliometrix software [31]. In this review paper, relevant papers were selected from the search results of Scopus and Google Scholar databases to discuss critical findings reported by researchers under the sub-topics in the next section.

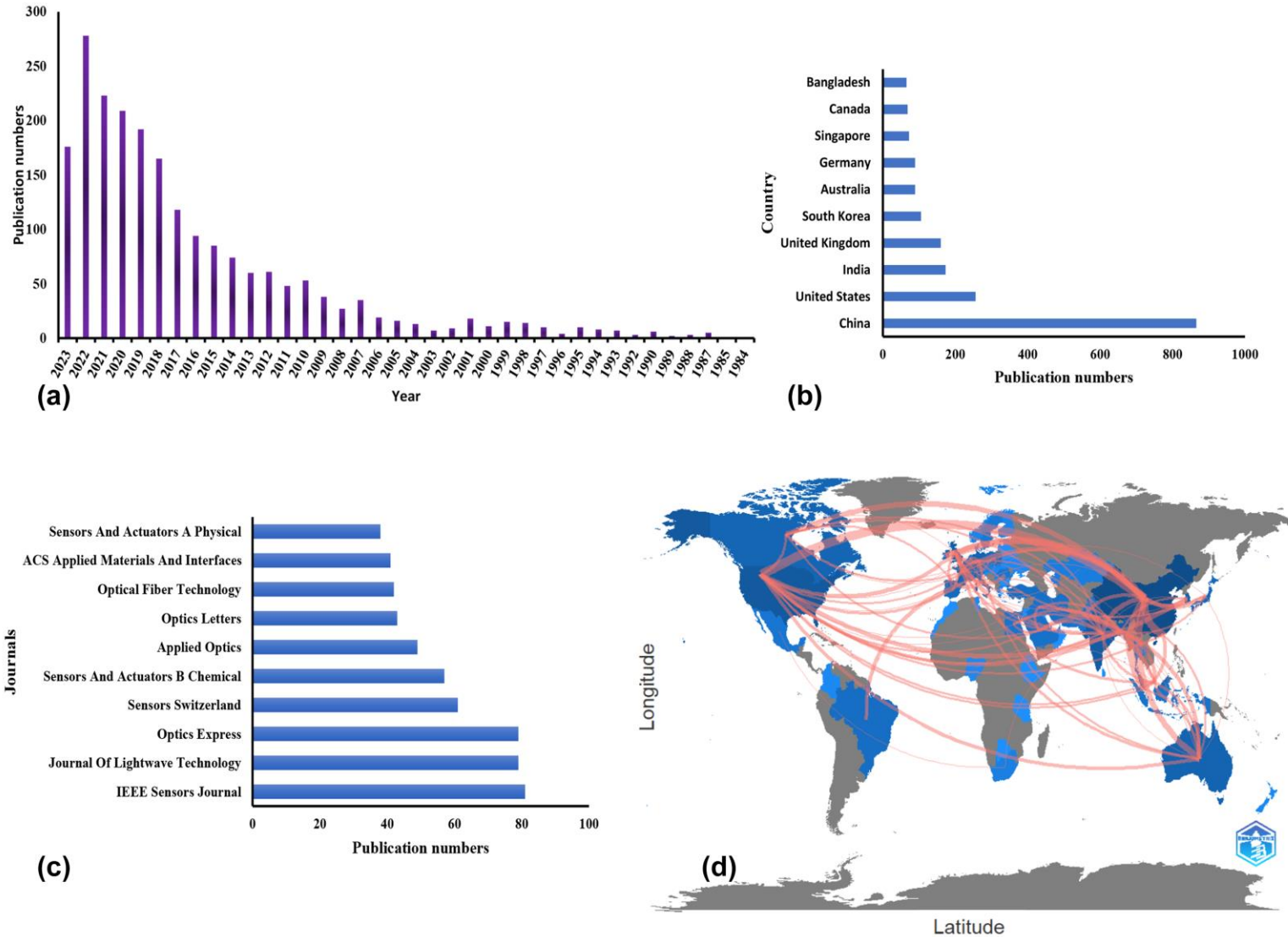


Figure 1. (a) Number of publications in recent decades (Data obtained from Scopus using the search blocks "fiber-based" AND "sensors" OR "supercapacitors" OR "energy-autonomous systems"), (b) Top contributing countries, (c) Top leading publishing journals and (d) Visualization of collaborations from countries in the World.

### 3. Fiber-based supercapacitors

Fiber-based SCs are flexible materials with great electrical and mechanical properties that can easily store energy in their electrodes [32]. With the ability of SCs to provide long cycle life and high power density, these fiber-based SCs are commonly produced using four ideal materials; namely carbon-based materials, metal oxide nanoparticles, conductive polymers, and their composites as electrodes [21]. Table 1. details the features of common material electrodes used in fabricating fiber-based SCs.

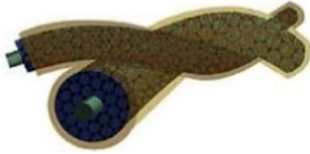
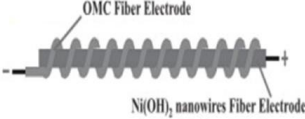


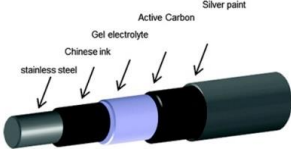
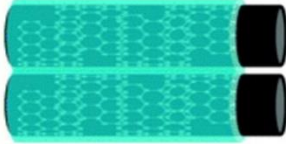
Table 1. Characteristics of material electrodes for SCs [21].

<b>Material electrode</b>	<b>Strengths</b>	<b>Weakness</b>
Conductive polymers	High stretchability, flexibility, conductivity, and faradaic capacitance.	Low energy and power density
Carbon based materials	Large surface area, low mass density, long cycle life, good stretchability and chemical stability, high conductivity.	Very costly High energy density High faradaic capacitance
Metal oxides	High energy density and faradaic capacitance.	Low stretchability and flexibility

Fibers used in the development of SCs have several characteristics that make them suitable for energy storage applications due to their high surface area which allows a greater amount of charge to be stored per unit volume. Also, the porous nature of fibers allows for the diffusion of ions and electrolytes, which is important for the operation of supercapacitors. Fiber-based SCs allow for the flow of electrons and ions necessary for energy storage resulting in EAS that can conform to different shapes and sizes possessing high tensile strength. These characteristics of fibers make them ideal materials for the development of SCs with high energy density and power output.

Fiber-based SCs are designed using four structural configurations, namely; coaxial, twisted, consecutive and parallel as encapsulation layers, electrolytes, or electrodes [33]. Table 2 summarizes the different configurations used in fabricating fiber-based supercapacitors.

Table 2. Description of configurations used in fabricating fiber-based supercapacitors.

Configurations	Diagram	Description	Ref.
Two-ply		A gel polymer electrolyte is twisted together with two fiber electrodes. This gel polymer acts as a separator in-between the fibers.	[34]
Coaxial helix		A stable fiber electrode is wound with a fiber electrode in a coil-like manner with a gel polymer electrolyte or separator.	[35]
All-in-one		The fabrication process integrates the separator and the two electrodes into a single fiber with limited extra bonding.	[36]
Coaxial stretchable		Elastic fiber with gel electrolyte is wrapped with electrode materials. The use of elastic fiber helps promote stretchability.	[37]
Coaxial		An outer layer electrode and a core fiber electrode with a gel polymer electrolyte or separator in-between the fibers.	[38]
Parallel		Two fiber electrodes are closely fabricated in a parallel manner with a gel polymer electrolyte or separator.	[39]

Fiber-based SCs are generally categorized based on the performance and selection of the electrode materials, for electrochemical double-layer capacitors (EDLC), pseudo capacitors and hybrid SCs [40]. SCs have a working principle of electrostatically storing charges at the interface of electrode and electrolyte material (Figure 2). SCs commonly consist of parts, an electrolyte, two electrodes and a separator to ensure its functionality and performance. These porous electrodes are immersed within the electrolyte solution with a thin separator membrane that prevents contact but ensures the passage of ions [41]. An applied voltage causes the ions in the electrolyte to move closer to the opposite charge unit of the electrode materials, a situation that

forms a charge layer at each electrode. This results in one of the electrodes being positively charged and the other being negatively charged. Because of this activity, an electrical double layer is formed with an accumulated electric charge on the electrodes [42]. Alternatively, the anion (negatively charged ions) and the cation (positively charged ions) are released from the surface of the electrodes and return to the electrolyte during discharge [43].

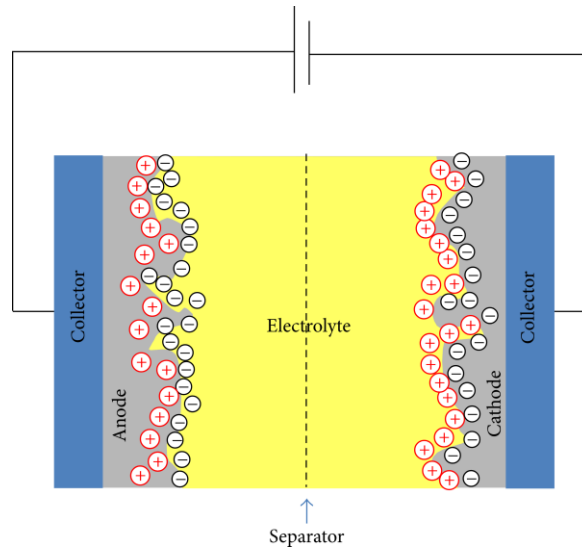


Figure 2. Schematic diagram of SCs working principle [44]. (An open access article distributed under the Creative Commons Attribution License)

### 3.1 Electrochemical double layer capacitors

The energy storage in EDLCs is primarily based on the electrostatic double-layer formed at the interface between the electrode and electrolyte as shown in Figure 3. The structure comprises an electrolyte, separator and electrodes. In EDLCs, the electrostatic interaction occurs at the interface of electrode/electrolyte and the needed energy is stored from charged ions when there is an applied external potential on the electrode [45]. This results in no ion exchange between the electrolyte and the electrode but rather a purely physical absorption [46]. Furthermore, the appropriate energy density is achieved in EDLC when there is an increase in the specific surface area due to the electrode distance decrease from the integration of the double layer. EDLCs are fabricated using electrode materials such as carbon nanotubes, graphene, and activated carbon materials, largely due to their excellent conductivity, chemical and thermal stability, and large specific surface area [45]. The key benefits of EDLCs are their high-power density, fast charge and discharge capabilities, great cycling stability, and extended cycle life.

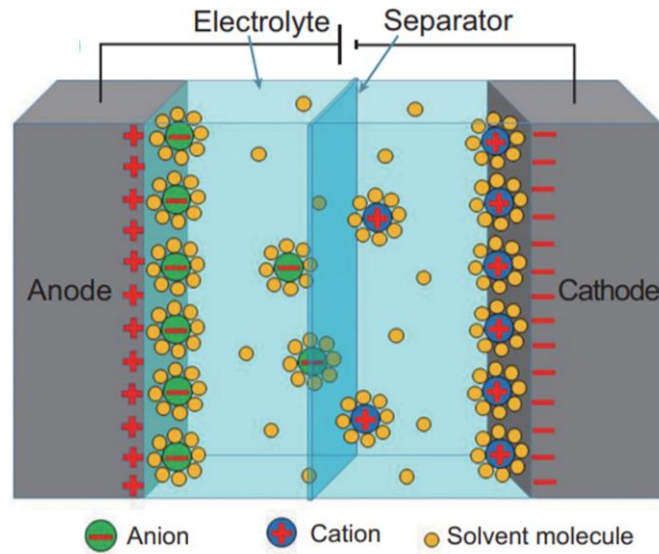


Figure 3. Electrochemical double layer capacitors [47]. (An open access article distributed under the terms of the Creative Commons CC BY license).

They can produce bursts of high power when needed and withstand numerous charge-discharge cycles without deterioration. However, as compared to batteries, EDLCs usually have a lower energy density, limiting their overall energy storage capacity [48]. EDLCs are used in a variety of applications, including automotive systems, renewable energy storage, power backup systems, portable electronics, and regenerative braking. Ongoing research intends to improve EDLCs' energy density, operating voltage, and overall performance in order to broaden their variety of uses in the future.

### 3.2 Pseudo capacitors

In this type of SCs, the pseudo capacitance is caused by reversible faradaic redox reactions at the electrode/electrolyte contact [49]. Pseudo capacitance is the reversible transport of electrons between the electrode surface and the electrolyte as shown in Figure 4. This mechanism is due to the reversible redox reactions of particular materials, such as transition metal oxides or conducting polymers, which may store charge via surface-bound or intercalation processes. Pseudo capacitors add greater capacitance values than regular capacitors and enable extra energy storage beyond the EDLC method [46].

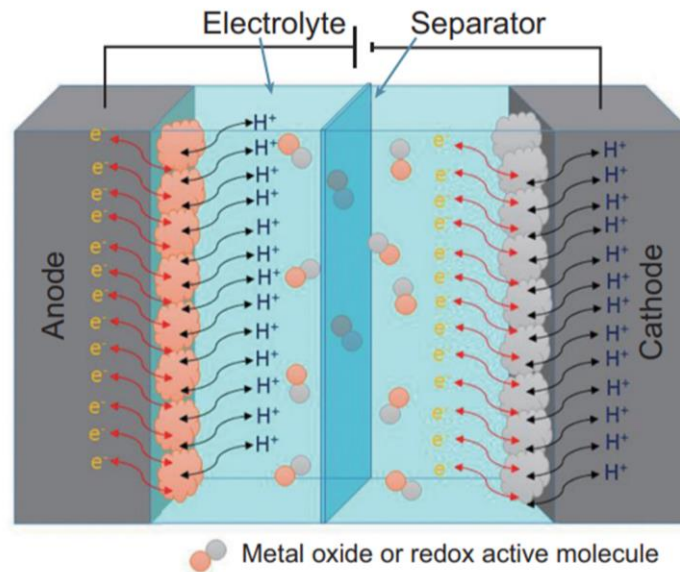


Figure 4. Pseudo capacitors [47] (An open access article distributed under the terms of the Creative Commons CC BY license).

This category of SCs has a short cycle life, less electrolyte accessibility and poor conductivity [50]. Alternatively, pseudo capacitors have a high energy density, wide operating temperature range, and specific capacitance due to the faradic process. Because of these benefits, pseudo capacitors are a viable energy storage technology for a wide range of applications, including portable electronics, transportation, renewable energy integration, and grid-level energy storage. There is ongoing research and development efforts to increase their performance, energy density, and cost-effectiveness, propelling their acceptance in a variety of sectors.

### 3.3 Hybrid supercapacitor

Hybrid supercapacitors (HSCs) combine the power source of EDLC and the energy source of pseudo capacitors for improved properties and characteristics as shown in Figure 5 [49]. HSCs leverage on the high-power density and quick charge-discharge capabilities EDLCs as well as batteries' high energy density. The HSCs are made up of three major components, namely EDLC electrodes, pseudo-type electrodes, and a compatible electrolyte system [51]. One electrode in the HSC is built using EDLC principles and is often made of a high-surface-area material such as activated carbon or carbon nanotubes, allowing effective charge storage via the electrical double-layer mechanism.

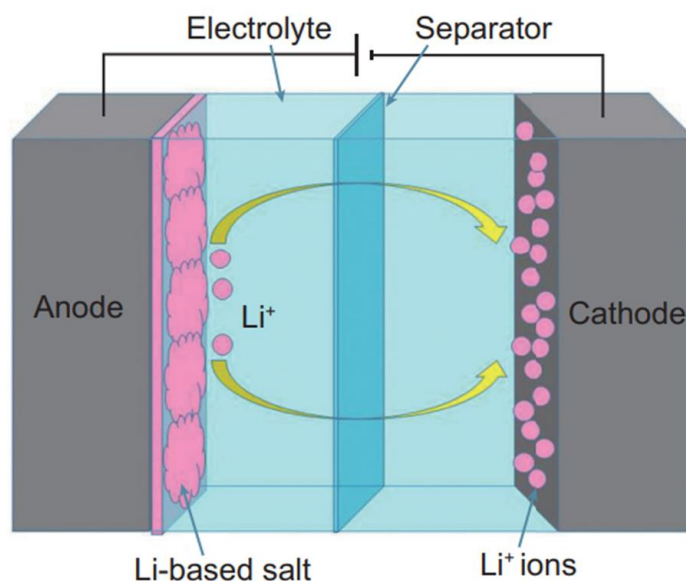


Figure 5. Hybrid SCs [47] (An open access article distributed under the terms of the Creative Commons CC BY license).

The EDLC electrode offers SCs' high-power density and quick charge-discharge characteristics. The HSC's other electrode is made of pseudo-type electrode materials. Compared to EDLC electrodes, these materials can undergo reversible redox reactions, allowing for greater energy storage. Transition metal oxides and conductive polymers are common pseudo-type electrode materials used in HSCs [52]. The increased energy density of the pseudo-type electrode allows for better total energy storage capacity. Also in the HSC system architecture component is an electrolyte that is compatible with both EDLC and pseudo-type electrodes. The electrolyte used is determined by standards such as the operating voltage range sought, temperature stability, and electrode compatibility. These SCs exhibit the properties of both materials for higher power and energy densities. Their improved performance makes them more favorable for energy-efficient systems due to the mechanism of energy storage [49]. HSCs are used in a variety of sectors where a balance of power and energy is desired. They are used in hybrid and electric cars, renewable energy systems, grid energy storage, and other applications that demand high power output as well as long-term energy storage [49].

#### 4. Fiber-based sensors

In this section, it is important to establish the foundation by providing details on general working principle of sensors before proceeding to present findings from studies on fiber-based sensors. Sensors generally convert physical and chemical variables of a process or an installation into

electrical signals that almost always begin as analogical signals [53]. This conversion usually mirrors as closely as possible the variables involved. While specific sensor types can vary in their working principles depending on the intended application and the measured parameters, the general working principles of sensors involve a transduction mechanism that converts the physical, chemical, or biological quantity being measured into an electrical or optical signal [54]. Transduction occurs through various mechanisms such as changes in resistance, capacitance, inductance, voltage, current, optical properties, or other measurable parameters. The transducers are connected to a sensing element that interacts with the target parameter and produces measurable responses based on different principles, including mechanical, electrical, magnetic, optical, or thermal properties. The responses are detected using different mechanisms to capture and measure the response from the sensing element, which can involve measuring changes in electrical signals, optical properties, or other physical parameters [55]. This process is followed by signal processing. Signal processing techniques may be applied to enhance the accuracy, reliability, or interpretation of the obtained data. This can involve amplification, filtering, linearization, calibration, or other signal conditioning techniques [56]. Also, all sensors provide an output signal that represents the measured parameter. The output can be in various forms, such as analog voltage or current, digital data, frequency, or optical signals. The output is typically compatible with the interface requirements of the system or device to which the sensor is connected. As a general principle, all sensors require calibration to ensure accurate and reliable measurements. Calibration involves comparing the sensor's output to known reference values or standards to determine any systematic errors and applying correction factors [57]. Calibrated sensors provide accurate and traceable measurements within specified ranges. Again, all sensors have specific sensitivity and measurement ranges that define their ability to detect and measure the target parameter. Sensitivity refers to the minimum detectable change in the measured quantity [58]. The measurement range defines the upper and lower limits of the parameter that the sensor can accurately measure.

Fiber-based sensors are generally fabricated to detect environmental and physiological changes in real-time [21]. Due to their inherent functionality for detecting changes in both the body and environment, they can be easily integrated into textiles for advances in EASs. These autonomous or self-powered or energy-autonomous systems rely on energy generators, EES devices, wireless power technologies, self-powered sensors, and hybrid energy systems combining both energy generators and EES [59, 60]. For applications like lighting LEDs and other sensing capabilities, fiber-based SC in series or parallel configuration are ideal;

nevertheless, their concurrent mechanical stability and energy replenishment might be difficult at times. For instance, while energy can be restored in a normal sweat-based SC, the mechanical stability of the material employed is a concern that might impair performance, especially during washing. To overcome such obvious challenges, autonomous system design structure requires an energy storage unit that stores energy harvested from nanogenerators and provides the required power to operate the functioning units such as transceivers and sensors [61].

The main problem with autonomous systems is that the power and voltage output that energy harvesters normally provide might be less than what is needed to charge SCs and regular batteries (which store energy) to operate a device. Therefore, feasible approaches include; shrinking systems that store energy, reducing the power consumption of the device and integrating the different components of the system [62].

For fiber-based SCs meant for EAS, they are mostly fabricated for use as temperature, photo, chemical and tactile sensors using different applications, materials and methods. In terms of size, due to the micro nature of fibers, micro-SCs ( $\mu$  SCs) are required to have the ability to store and deliver power in short times to meet demand. M SCs are different from  $\mu$ -batteries due to their superior cycling stability and power peaks for important applications. For example, from an electrostatic process, charge energy is stored and delivered from carbon electrodes having a high surface area on fiber-embedded electrochemical double-layer capacitors (EDLCs). For pseudo-supercapacitors, metal oxides and conducting polymers are used for battery-like electrodes which can be processed into fibers through any of the methods in Table 3. These are through reversible and fast redox processes charged and discharged [12]. Subsequently, through several faradic and electrostatic modes, electrode materials (of negative and positive) are charged and discharged for hybrid supercapacitors. Due to the flexible architecture of fibers and ease of processing, polymers and carbons are ideal materials used to fabricate  $\mu$  SCs.

Table 3. Summary of studies on the application of fiber-based sensors [21]

<b>Application</b>	<b>Materials used</b>	<b>Sensing target</b>	<b>Methods of fabrication</b>	<b>Sensing and Mechanical properties</b>	<b>Ref.</b>
Temperature fiber-based sensor	Single reduction graphene oxide (rGO)	Temperature for biomedical and healthcare monitoring.	Wet spinning	It showed good response and recovery time of 7s and 20s respectively, and responsivity to change in temperature (from 30° to 80°C). Even at 10 000 bending cycles and 4 mm bending radius, the response rate was still good (see Fig 1A)	[63]
	chalcogenide semiconductor (Ge <sub>17</sub> As <sub>23</sub> Se <sub>14</sub> Te <sub>46</sub> )	Thermal sensing	Thermal drawing process (TDP)	The fiber sensors are flexible and lightweight Sensing ranges from 11°C to 58°C (see Fig 1B)	[64]
Humidity	Single-walled carbon nanotubes (SWCNTs)-poly vinyl alcohol (PVA) filaments	Humidity	Wet spinning	Short response time of 40s. A more than 24 times increase in electrical resistance under a high RH of 100% as compared to under a low RH of 60%.	[65]
	cotton threads coated with carbon nanotubes (CNT) and polyelectrolytes	Humidity and albumin detection	Dipping technique	The network of nanotubes of 20Ω/cm promoted efficient charge transport	[66]
Tactile	Conductive fibers coated with dielectric	Pressure	Coating	High stability after 3 000 bending tests, quick response time and high sensitivity of 0.21 kPa <sup>-1</sup> , fast relaxation time of less	[67]

	rubber materials			than 10 ms, high durability after 10 000 cycles, electrical property of $0.15 \Omega \text{ cm}^{-1}$ .	
	Cotton fibers and Silver-coated nylon fiber	Strain	Core spinning	15% strain test resulted in a gauge factor of 0.695. Mechanical property of 10,000 cycles representing high stability	[68]
Photo-sensor	Cellulose based thread-single walled carbon-nanotubes (SWCNTs)	UV monitoring	Dip-coating	Good mechanical property after 500 bending cycles, good response to UV rays.	[69]
	p-CuZnS/n-TiO <sub>2</sub> with CNTs	UV monitoring	Deposition	Good responsivity of $640 \text{ A W}^{-1}$ Ultrahigh photocurrent of $\approx 4 \text{ mA}$ Faster response rate.	[70]
	Amorphous chalcogenide glass (As <sub>40</sub> Se <sub>60</sub> /As <sub>40</sub> Se <sub>54</sub> Te <sub>6</sub> )	Imaging	Thermal drawing process	Resolution of $4^\circ$ angular and 5 nm wavelength.	[71]
Chemical sensor	PEDOT:PSS	Sodium chloride	PEDOT:PSS	Very sensitive at $600 \mu\text{A/dec}$ , good mechanical property after 1000 bending cycles	[72]
	Gold fiber modified with Ag/AgCl	Glucose	Dry-spinning, coating, electrodeposition	$11.7 \mu\text{A mM}^{-1} \text{ cm}^{-2}$ sensitivity 0–500 $\mu\text{M}$ linear range Mechanical property of 1000 releasing and stretching cycles 200% strain electrochemical properties	[73]

#### 4.1 Temperature sensing

Fiber-based sensors are fabricated to measure temperature changes in the body from 35°C to 38°C providing the person’s health state as well as indicating other pulmonological diagnostics and cognitive state [74, 75]. These temperature sensing fibers are made of electrodes that employ a working principle of using changes in the electrical resistance of the electrode material to measure the temperature changes in the environment. These fiber-based sensors are typically flexible electrodes based on two mechanisms (thermos-resistive and thermos-electric) which are in contact with heat-sensitive material [21]. Due to their flexible nature, they are easily integrated to form textiles, which come into contact with the human skin directly hence facilitating the continuous monitoring of changes in skin temperature due to interaction with the environment [63]. Critical examples of these research advances in fiber-based temperature sensors are illustrated in Figures 6 (a-f).

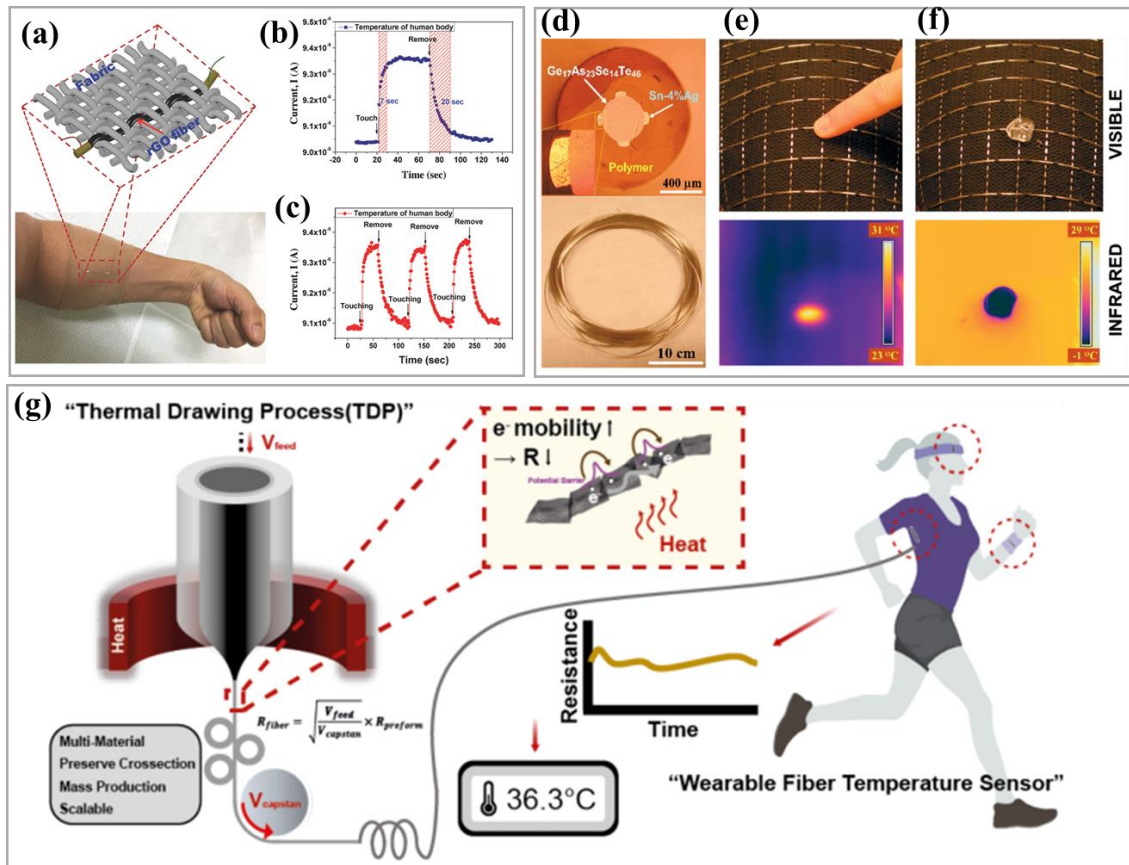


Figure 6. (a) Image of an arm wrapped with the fiber-based temperature sensor fabric, (b) Results showing a 7s fast response time and 20s quick recovery time, (c) Time-dependent response current under three cycles of exposure to the human body. Adapted with permission from Ref. [63]. Copyright 2018 John Wiley and Sons. (d) Cross-section of the thermal sensing fiber (e) Heating due to touch on the constructed fabric and (f) Cooling with an ice cube. Adapted with permission from Ref. [64]. Copyright 2006 John Wiley and Sons. (g) Temperature sensing fiber [76]. (This article is licensed under a Creative Commons

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In a recent study conducted [76], a temperature fiber-based sensor (Figure 6g) was fabricated using a linear flexible low-density polyethylene layer (for the outer core) and, conductive polymer nanocomposite (thermoplastic polylactic acid), reduced graphene oxide and a conductive filler (for the inner core). The manufacturing process used a thermal drawing technique to create this multi-material temperature fiber-based sensor. Findings revealed that, with a 14.8 second recovery and a 11.6 second rapid response time, the fiber-based sensor could measure 25 to 45 °C temperature range with good sensitivity.

#### ***4.2 Humidity sensing***

The effects of dry and moist air on the skin vary from one person to another based on their geographical location. The working principles of humidity sensors are based on the change in electrical resistance largely due to the desorption or adsorption of water molecules from the ambient air in the surroundings (temperature and relative humidity). There is changes in conductivity of the electrodes in the sensors when water is absorbed causing a dissociation of the ionic functional groups, hence material resistance decreases [77]. Several research studies have been conducted in an attempt to develop a humidity sensor with high sensitivity for effective humidity monitoring of human health. These sensors can easily monitor and measure the presence and amount of water or water vapor in the surrounding air in the environment and provide feedback to the wearer in many ways. The use of flexible fiber-based supercapacitor humidity sensors promotes the possibility of fabric formation which can be used in sensing for effective humidity monitoring.

The use of flexible fiber-based SCs has received attention due to the long cycling stability, high power density, rapid rate of charge and discharge, and impressive stretchability. Despite the impressive characteristics of flexible fiber-based supercapacitor humidity sensors, they are thus limited for wearable and flexible applications due to their relatively low energy density. In light of the low energy density common with FFHS, [78] fabricated a 3D fiber structure with a nickel cone (highly conductive) and an active substance (metal oxide-  $\text{MnO}_2$ ). Here, the Nickel cone is grown on carbon nanotubes (CNT) where  $\text{MnO}_2$  is further electrodeposited. This cost-effective and facile approach provided an asymmetric supercapacitor with a high operating voltage window, high energy density, and  $0.5 \text{ mA/cm}^2$  of current density with a  $609.06 \text{ mF cm}^{-2}$  of high specific capacitance.

The finished flexible fiber-based supercapacitor humidity sensors device exhibited an

outstanding areal energy density of  $83.59 \mu\text{Wh cm}^2$ , a high specific capacity of  $195.38 \text{ mF cm}^2$ , a flexible humidity sensor sensitivity of  $2.483/\% \text{ RH}$  in the detection of relative humidity (RH), and a quick response time of  $0.39 \text{ s}$  [78] as shown in Figure 7. To advance the concept of sustainability into flexible fiber-based supercapacitor humidity sensing, Mankkajal et al [20] fabricated natural fiber-based sensors (humidity and temperature) and SC. Here, the separator is the cellulose material (jute fiber) and the electrode is formed by coating PEDOT: PSS/SWCNT on the surface of the jute fiber. The temperature sensor had a  $0.23\% \text{ }^\circ\text{C}^{-1}$  change in response (from  $24^\circ\text{C}$  to  $35^\circ\text{C}$ ) while a  $1.5\Omega/\% \text{ RH}$  sensitivity was recorded for the humidity sensor.

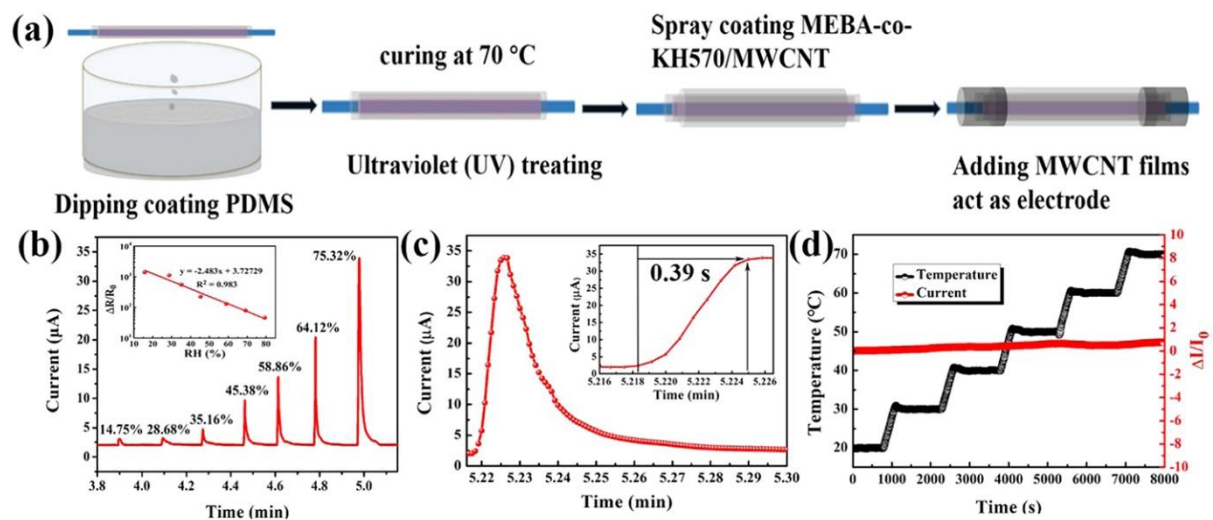


Figure 7. (a) Schematic illustration of the integrated humidity sensor, (b) MEBA-co-KH570/MWCNTs-sensitive composite-based sensor real-time dynamic response curve with relative humidity ranging from 14.75% to 75.32%, (c) Sensor exhibited a fast response time approximately 0.39 s, (d) The effect of varying temperatures on the humidity detection capabilities of the MEBA-co-KH570/MWCNTs based sensor tested across a range of  $20^\circ\text{C}$  to  $70^\circ\text{C}$ . Adapted with permission from Ref. [78]. Copyright 2019 Elsevier.

### 4.3 Tactile sensing

Touch perception is a fundamental survival ability of humans, and developments in tactile sensor technology have been implemented in a variety of sectors, offering advantages including exceptional item manipulation and broad human-robot interactions. The working principle of tactile sensors is governed by four ideal effects: triboelectric, piezoelectric, piezo capacitive and piezo resistive [79]. For piezoelectric effect, there are changes in the alignment of electric dipoles when a mechanical force deforms a crystalline solid creating an electric field inside [80]. For triboelectric effects, there is an electric charge when the electrode

materials come into contact with and are separated from the surface of another material [81]. For piezo capacitive effects, the capacitance changes when an external force changes the physical shape of the dielectric [79]. Lastly, for piezo resistive effects, mechanical strain influences the electrical resistance of semiconductor changes, where electron movements due to strain effects alter the conductivity [82]. Fiber-based sensors are fabricated for pressure or touch or force or strain sensing on the body due to their breathability and long-time use. Advances have been made on fiber-base sensors to replace conventional e-skins due to their longevity and breathability issues even though they are flexible and have good sensing abilities [83]. Furthermore, these fiber-based sensors help in detecting resistance, capacitance, pressure, and touch. The sensing mechanisms of fiber-based SCs can be grouped as capacitive, piezoelectric, and resistive sensors [17]. Any applied stimulus is determined by changes in capacitance. This is made possible with a shift in the geometry (area between the two electrodes) of the sensor. With a change in mechanical stimuli, piezoelectric sensors can convert that into electrical energy hence promoting self-powering abilities. Resistive sensors are used to determine the stress amplitude when there is a resistance change in mechanical stress. A most recent study fabricated a tactile sensor (Figure 8) using optical fibers with a 92.41% recognition accuracy for a 0 to 3.5N contact force [84]. A successful coaxial piezoelectric fiber based electronic skin was fabricated for tactile sensing [85]. Findings revealed the high sensitivity rate of the fiber for 80 to 230kPa pressure range.

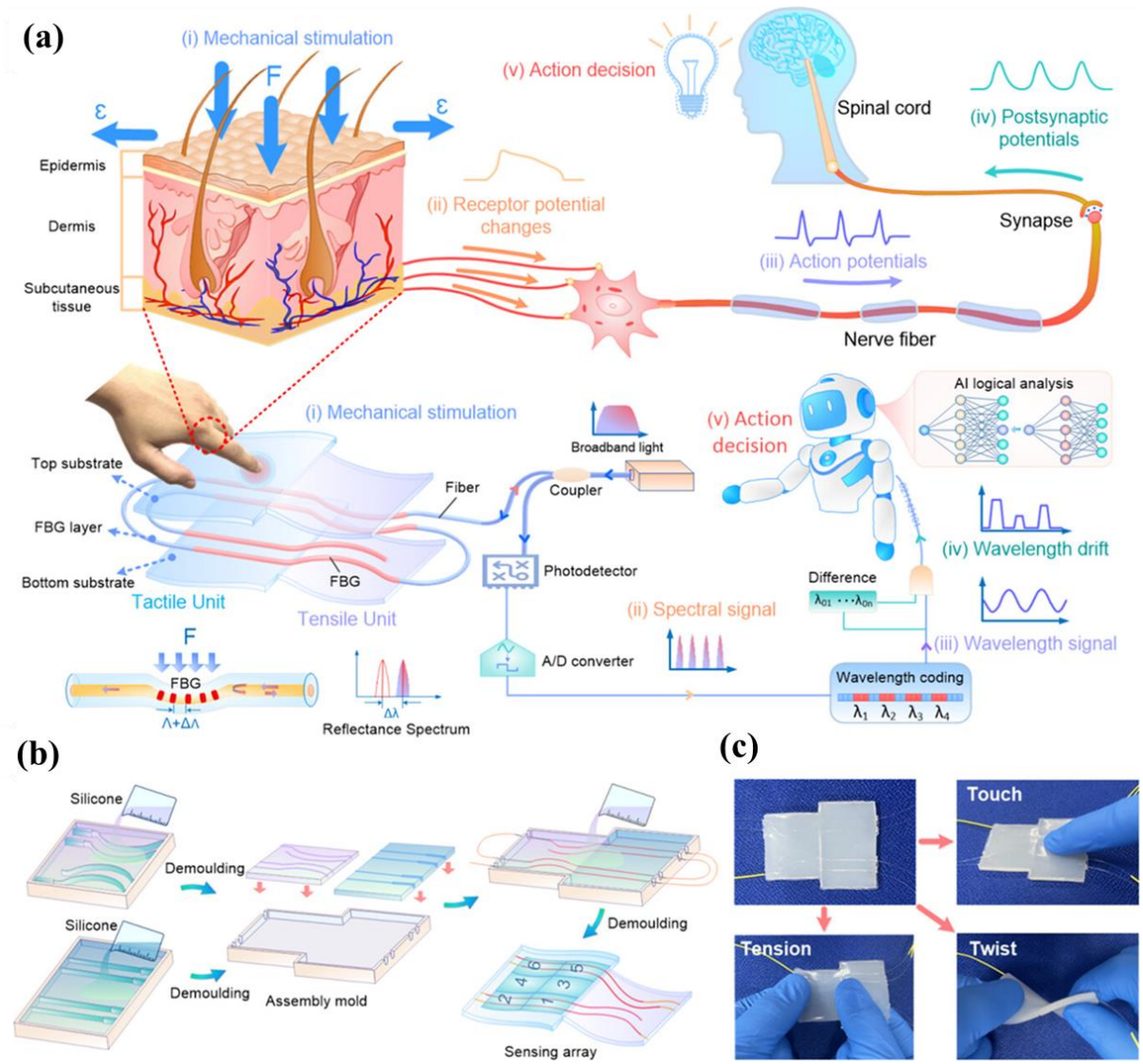


Figure 8. The fabrication process of the sensor. (a) Design inspiration and structure of the sensor, and artificial tactile cognitive systems that mimic biological systems, (b) Fabrication process of the sensor, (c) Physical properties of the Sensor. [84]. (This article is licensed under the terms of the Creative Commons Attribution License).

#### 4.4 Photo-sensing

Conventional photo-sensors are relatively rigid and require lenses for optical communication, ultraviolet radiation monitoring, scanning, and imaging [86]. The fabrication of fiber-based humidity sensors provides flexible materials that can be woven into a lightweight textile easily. This provides a great surface area for detecting the wavelengths and directions of the electromagnetic field. UV radiations are typically classified depending on the wavelength as UV-A (with wavelengths of 315 nm to 400 nm), UV-B (with wavelengths of 280 nm to 315 nm), and UV-C with wavelengths of 10 nm to 280 nm) [69]. Due to such inherent

functionality of the textiles, monitoring the extend of exposure of the skin to UV-A and portions of UV-B radiation is possible since excessive exposure leads to skin diseases like cancer, cataracts, and erythema when individuals stay in the sunlight for long hours [87]. A typical example of fiber-based photo-sensors is illustrated in Figure 9 (a-i).

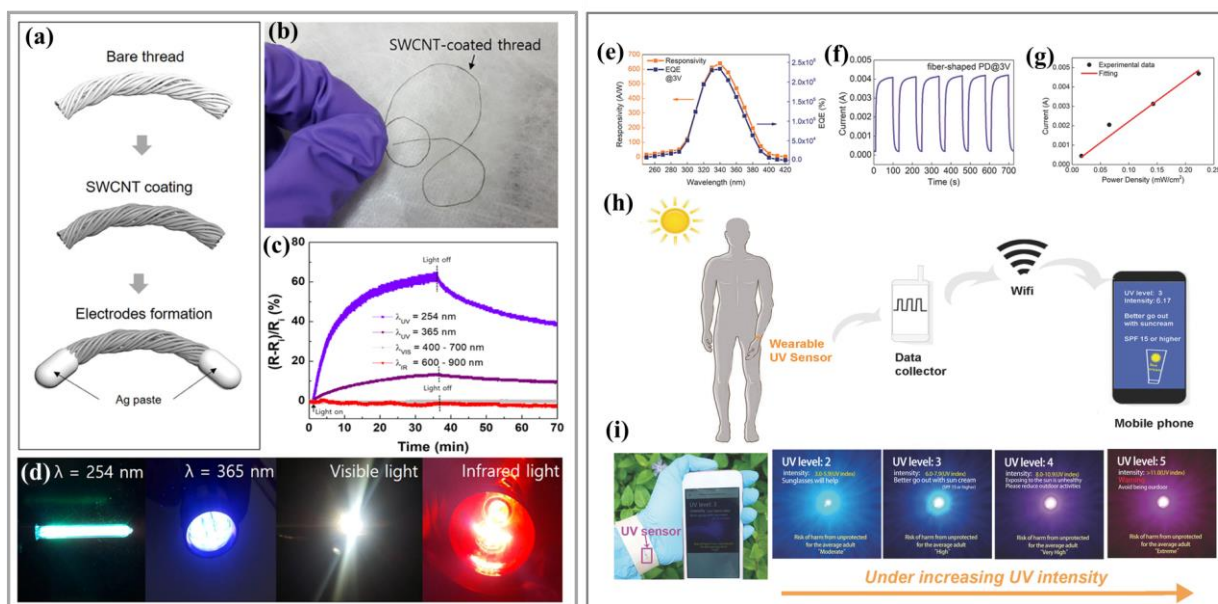


Figure 9. SWCNT-coated cotton thread. (a) Schematic illustration of the fabrication process of a SWCNT-coated cotton thread. (b) Photograph the SWCNT-coated cotton thread. (c) Relative resistance changes of the wearable UV sensor upon constant UV illumination. (d) Photographs of four different light sources. Adapted with permission from Ref. [69]. Copyright 2018 American Chemical Society. Fiber-based p-CuZnS/n-TiO<sub>2</sub> UV sensor. (e) Responsivity and EQE of the fiber-shaped UV PD as a function of wavelength at 3 V, (f) The on–off switching tests of the fiber-shaped PD at 3 V under 350 nm, (g) Experimental and calculated photocurrent as a function of power density of the fiber-shaped UV sensor, (h) Schematic illustration of the wearable PD as a real-time UV monitor; (i) The photographs of a wearable real-time UV sensor in real life. Adapted with permission from Ref. [70]. Copyright 2018 John Wiley and Sons.

#### 4.5 Chemical sensing

Fiber-based chemical sensors play a critical role in the real-time detection of various chemicals on the skin or surroundings. These aid in healthcare monitoring by the detection of chemical markers of respiration, saliva, tears and sweat [88-91], and detecting exposure to hazardous vapor in the environment like hydrogen chloride, ammonia and nitrogen dioxide [92-94]. The working principle is that, by measuring the electric current produced based on

the electrochemical reaction of the chemical and the electrode in the environment, the chemical concentration can be measured. According to Lee et al. [21], these sensors are commonly used in sportswear to detect and monitor the glucose, sodium ions, cortisol, and pH levels in sweat. They further opined that this is due to the reduced risk of contaminating the skin, breathability, and comfortability of the sensors. This has influenced studies to produce fiber-based chemical sensors to provide real-time continuous information for effective health care monitoring, most especially for chronic related diseases. A recent study by [73] fabricated a strain-insensitive and highly stretchable Ag/AgCl gold fiber-based sensor to monitor the glucose levels in sweat. Results revealed that with the strain from 0-200%, the fiber produced a strain-insensitive reproducible electrochemical performance. The fiber-based sensor further produced a sensitivity of  $11.7 \mu\text{A mM}^{-1} \text{cm}^{-2}$  and a linear range of 0-500  $\mu\text{M}$ . This showed promising potential applications in monitoring glucose levels in human sweat. This influenced further studies by [95] to fabricate a gold-based fiber sensor using a dry spinning method for monitoring lactate in sweat. A standard three-electrode system i.e., counter, reference and working electrodes was fabricated using elastomeric gold fibers, Ag/AgCl coated gold fibers, and Prussian blue (PB)/LOx/chitosan (CS) coated gold fibers respectively (Figure 10). These materials were integrated into a textile with results showing great lactate detection in sensitivity of  $14.6 \mu\text{A/mM cm}^2$  in artificial sweat and  $19.13 \mu\text{A/mM cm}^2$  in PBS, the linear range of 0 mM – 30 mM and stretchability up to 100%. This demonstrated a great potential application in healthcare lactate monitoring.

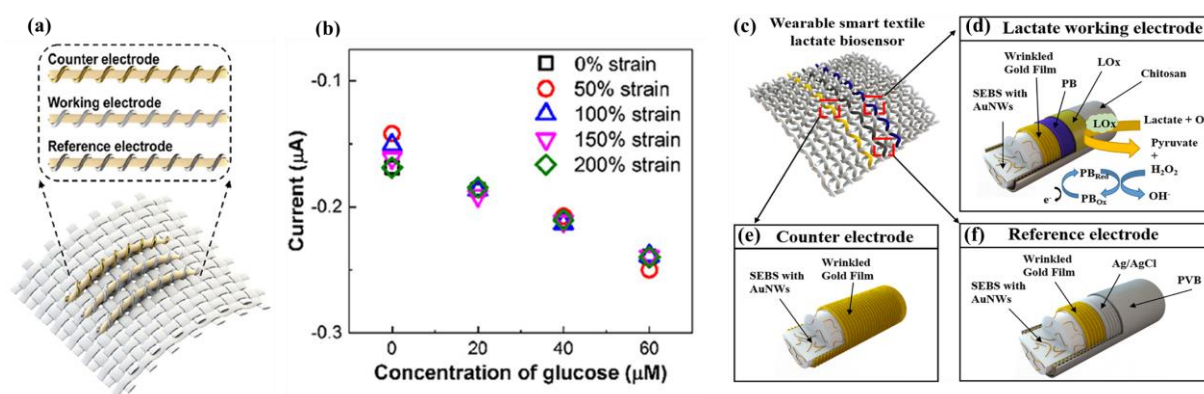


Figure 10. (a) Schematic illustration of the glucose sensors integrated into an elastic textile. (b) calibration plot of the chronoamperometric responses of the stretchable fiber-based glucose sensor from 0 to 200% strain. Adapted with permission from Ref. [73]. Copyright 2019 American Chemical Society. (c) Scheme of the woven textile biosensor toward lactate monitoring. (d) The morphology and mechanism of lactate working electrode. (e) The morphology of elastomeric gold fiber and it can act as a counter electrode directly. (f) The

morphology of PVB coated Au/Ag/AgCl reference electrode. Adapted with permission from Ref. [95]. Copyright 2021 Elsevier.

## **5. Methods of Fabrication fiber-based sensors and SCs**

Fiber-based sensors and SCs are fabricated using varying methods and materials for EAS. The use of these methods should be carefully selected to ensure the fabrication process does not affect the ultimate performance of the material. There are four main fabrication methods utilized for fiber-based sensors and supercapacitors, these include coating (dip-coating), spinning (wet, dry, melt and electrospinning), deposition (chemical vapor and electro depositions), and drawing (thermal drawing). Advanced studies in the field as opined by Lee et al. [21] utilize thermal drawing, deposition, spinning and dip coating in fabricating fibers for energy-autonomous systems. Table 4 provides a detailed description, of the advantages, and disadvantages of the fabrication methods used for fiber-based sensors and supercapacitors.

Table 4. Description of fabrication techniques used for fiber-based sensors and supercapacitors.

<b>Categories</b>	<b>Description</b>	<b>Advantages</b>	<b>Disadvantages</b>	<b>Reference</b>
Dip-coating	Substrates are dipped in solution with active materials to coat their surfaces. This could be done with the presence of a weighing roller which helps to form uniform coats on the surfaces of the substrates. Air drying follows immediately to cure the coatings.	This is a very fast and simple process that promotes mass production. Improved bonding between the electrochemically active materials and the substrate.	The coating thickness of the substrate's surface affects its adsorption and absorption. Active materials are consumed in large quantities. Sensitivity is low. Mechanical stability is poor.	[96-99]
Wet spinning	This process involves the extrusion of a mixture solution (made of active materials and polymers).	Mechanical stability is good.	Manufacturing time is long and complex	[21, 100]
Dry spinning	The process is similar to that of wet spinning, aside from the need for purification.	Productivity is high. Purification is not needed.	Mechanical stability is low. Toxic volatile solvents.	[21, 73]
Melt-spinning	The materials mixture is made viscous and extruded with the help of applied heat.	Solvents are not required. Low cost and simple process.	High temperature. Active materials have low concentrations.	[21, 101]
Electrospinning	The fibers are formed using electric force to pull the polymer solution	High mechanical stability	Requires high voltage	[21, 102, 103]
Electrodeposition	An electric field is used to deposit a solution of colloidal particles onto a conductive material	Low cost. High conductivity.	Time-consuming production process. Mechanical stability is poor.	[104]

Chemical Vapor Deposition	This process involves growing chemically active fine particles by decomposing and depositing gas reactants using plasma energy. Hydrocarbon substances are the main component used in the process, with nickel, cobalt, and iron used as a catalyst.	Sensitivity is high. Bonding on the substrate is strong.	Uncontrollable thickness of the substrate.	[97, 105]
Thermal drawing	With the application of heat, fibers are fabricated from a perform that incorporates different materials.	High productivity and sensitivity. Fiber thickness can be adjusted	Limited materials	[106]

## 6. Natural fiber-based composite SCs and sensors

Natural fibers are raw materials sourced from nature with appropriate properties like reduced weight, high strength, and bio-degradable [107]. They are further fibrous and flexible in nature with varying microfibrils and fibril structures. These structures ensure strong adsorption abilities to polar solvents and water. Such properties have advanced its use for SC and sensors for textile or woven fabrics. Due to the insulating nature of natural fibers especially cellulose as opined by Yadav [33], sufficient conductivity must be achieved during the fabrication process to ensure the best performance and functionality.

The use of natural fibers has become relevant in recent times for the fabrication of SCs and their application as sensors for eco-friendly devices. These are prominent because of the use of Li-ion batteries identified as regular energy storage [108, 109] which produces electronic waste largely due to the use of unfriendly materials [110]. Such unfriendly materials pose serious environmental threats even though they offer reliable output power and effective energy density. As such, natural fibers have been fabricated in recent times for improved SCs(SCs) and sensors suitable for energy autonomous systems, which are sustainable friendly materials and can be discarded naturally. This was demonstrated in a study conducted by Manjakkal et al. [20], where a jute-based fiber SC and sensors (for humidity and temperature) were fabricated. In the design of SC (Figure 11a-c), the jute fiber is first coated with poly(3,4-ethylene dioxythiophene) polystyrene sulfonate (PEDOT:PSS) and then functionalized with a single-walled carbon nanotube (SWCNT) on top. This approach offers chemical stability, excellent electrochemical activity, large surface and high aspect ratio, and electrical conductivity[111]. In the design of the sensors, ethylene glycol (EG) was mixed with the PEDOT:PSS and drop cast on the jute fiber. The fabrication of this electrode produced temperature (which was later covered with cling film) and humidity sensors. The addition of cling film aided in preventing humidity influences on the electrode. The design of the jute-based SC and sensors was further applied in a jute bag attached to a solar cell, where the sensors are operated by the SC using the stored energy from the solar cell. Results showed that at  $8.65 \text{ mF cm}^{-1}$  capacitance and  $0.1 \text{ mA}$  applied current, the power and energy densities of SC are  $3.85 \text{ } \mu\text{W cm}^{-1}$  and  $0.712 \text{ } \mu\text{Wh cm}^{-1}$ . This fabricated SC could

power the sensors. A sensitivity of  $1.5\Omega\%RH$  to  $50\%RH$  and a response of  $0.23\%^{\circ}C^{-1}$  were exhibited by the humidity and temperature sensors.

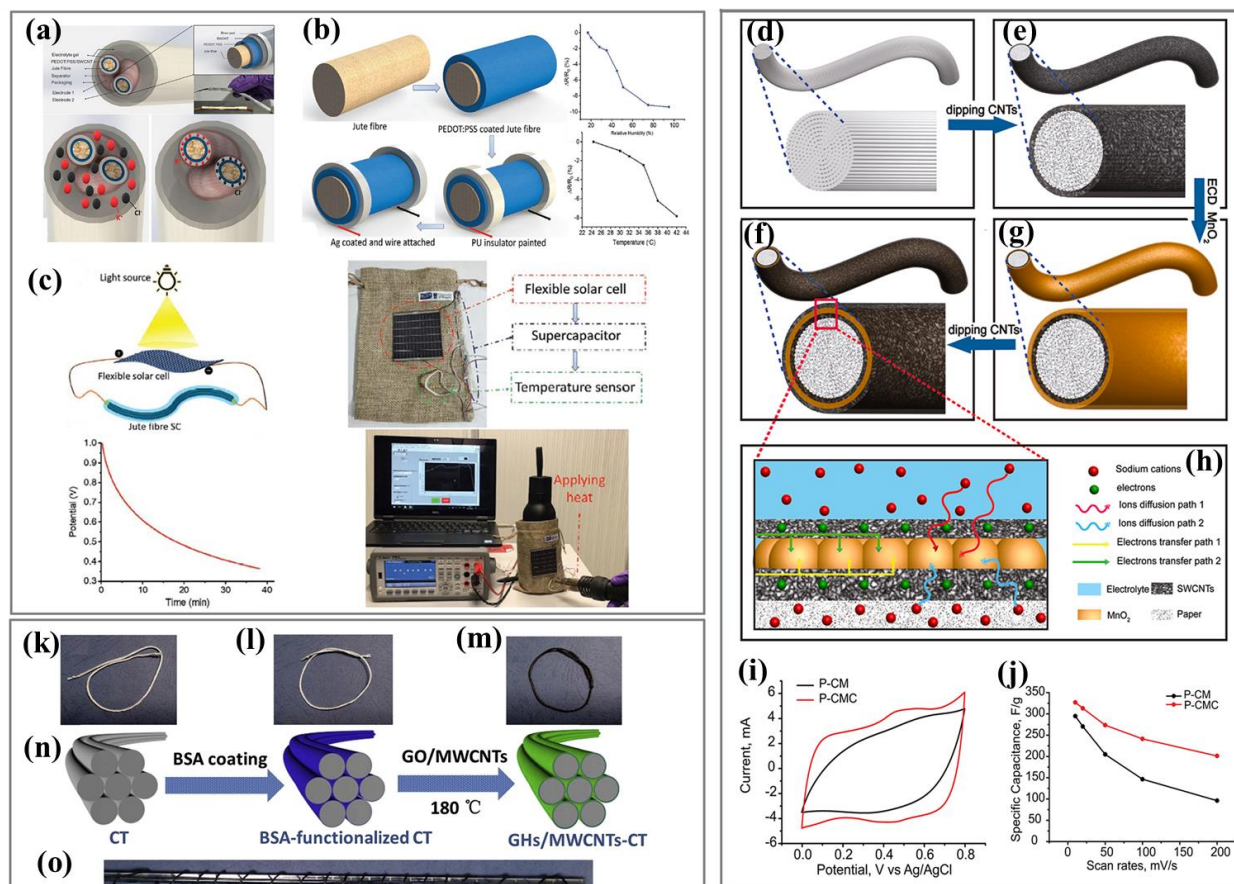


Figure 11. (a) Cross-sectional view of the fabrication of SC. (b) Cross-sectional view of the fabrication of jute fiber-PEDOT:PSS electrode for the humidity and temperature sensors coupled with their performance and (c) Integration of the jute fiber SC and sensors in a jute bag with flexible solar cells [20] (This article is licensed under the terms of the Creative Commons Attribution License - CC BY). Fabrication process and performance of natural cellulose fiber-based supercapacitors. (d) Image of one single cellulose fiber. (e) Dip-coating of CNT. (f) MnO<sub>2</sub> electrodeposition. (g) CNT's second dip-coating. (h) The magnified square area of the configured paper/CNTs/MnO<sub>2</sub>/CNTs (P-CMC) showing paths of ion diffusion and dual electron charge transfer. (i) P-CM and P-CMC cyclic voltammetry curves in 1 M Na<sub>2</sub>SO<sub>4</sub> electrolyte at 50 mV/s. (j) P-CM and P-CMC specific capacitances in a 1 M Na<sub>2</sub>SO<sub>4</sub> electrolyte at varying scan rates. Adapted with permission from Ref. [112]. Copyright 2018 American Chemical Society. (k-m) Images of k) CT, l) BSA-functionalized CT, m) GHs/MWCNTs-CT. (n) Schematic fabrication process of GHs/MWCNTs-CT. (o) a

50 cm long GHs/MWCNTs-CT wrapped around a 5.5 mm diameter glass rod. Adapted with permission from Ref. [113]. Copyright 2016 Elsevier.

With the mesoporous structures of cellulose fibers [114], Gui et al. [112] fabricated SCs using natural cellulose fibers for EAS. The fabrication process details the coating of carbon nanotubes (CNTs), manganese dioxide ( $\text{MnO}_2$ ), and another layer of CNT on a cellulose fiber (P-CMC) as shown in Figure 11d-j. The porous structure of the cellulose fibers acted as an electrolyte reservoir resulting in large electrolyte uptake and producing good electrochemical performance along the fiber. The coating of the second CNT was to optimize the SCs electrolyte by enhancing the delivering of electrons. Results revealed that such an addition further improved the electrochemical performance of the P-CMC configuration. This has dual electron and ion paths for improved capacitance and cycling performance. These results coupled with the mesoporous nature of cellulose fibers enhance good ion pathways which are critical for energy material deposition in energy storage. A most recent study, Wang et al [115] fabricated a multi-layer hierarchical conductive composite material using cellulose nanofibers (CNFs) / carbon nanotubes (CNTs) / vinasse activated carbon (VAC) (CCV) to prepare a supercapacitor with electromagnetic interference shielding (EMI) effect. The CVV composite materials exhibited excellent performance as SC and a high EMI efficiency. Other natural fibers like cotton have been fabricated and reported by Zhou et al. [113] in a study with graphene oxide (GO)/multi-walled carbon nanotubes (MWCNTs) mixture through a hydrothermal process (Figure 11 l-o). This effectively reduced the GO into graphene hydrogel (GH) with MWCNTs forming on the surfaces of the natural cotton fibers. The latter was first pre-treated with Bovine serum albumin (BSA) solution at room temperature. Due to the porous nature of the cotton fibrils, the assembly of the electrodes promoted the absorption and accumulation of ions for enhanced capacitance performance. A situation that is evident at the charge and discharge process hence promotes its application for energy systems. Further results revealed that due to the flexibility a high mechanical strength of the fabricated electrodes for SC, it can be used for knitted structures without affecting its electrochemical performance.

## **7. Synthetic fiber-based composites SCs and sensors**

Synthetic fibers and their composites present unique advantages for the fabrication of SCs due to their easily tunable properties during the process of manufacturing. Several studies have proven that [116-118]. These properties stem from the fact that synthetic fibers can be easily modified. The properties of the fibers are mostly dependent on the type of fiber used and the manufacturing process. In a study conducted by Zhao et al, it has been shown that synthetic fiber blend with nano particles aids the fabrication of high-performance SCs for sensing and various EAS applications [119]. Advantages of these fibers include high tensile strength, flexural properties, and high capacitance level. However, issues such as complicated fabrication processes with its attendant challenges such as high sensitivity to moisture, difficulty in scaling up, high-temperature processing (depending on the specific polymer) and expensive precursors remain [120]. Synthetic fibers such as carbon, glass and steel fibers are used extensively in the fabrication of SCs for sensors because of their lightweight and easy modification capabilities in areas of electric, optical, thermal, mechanical and chemical properties [121, 122]. The congenital versatility and volume of synthetic fiber-based materials can be transformed into a variety of shapes and structures, which in turn supports its integration with compact futuristic wearable electronic devices and gadgets. Below are some synthetic fibers with unique functionality mainly used for SCs meant for EAS applications.

### **7.1 Conducting polymer fiber-based SCs and sensors**

Conducting polymers (CP) are polymers that have the inherent property of conducting charge and hence are termed electrically conductive materials [123, 124]. The latter is due to CP containing functional groups with pseudo-capacitance characteristics [124]. Some examples of the conducting polymers used for fiber-based SCs include other polythiophene (PTh) derivatives, polyfuran (PF), poly(3,4-ethylene dioxythiophene) (PEDOT), poly(p-phenylene-vinylene) (PPV), polypyrrole (PPY), polyaniline (PANI) and polyacetylene (PA), shown in Figure 12a. These conductive polymer materials can easily be processed and stores charge through rapid faradic charge transfer [125, 126]. Aside from these,

Poly(ethylene glycol) (PEG) have been used to fabricate molecular crowding electrolyte for SC with a 2.5V performance [127]. Most CP are stretchable, flexible, conductive and have high faradaic capacitance even though they have relatively low energy and power density [21]. However, CP has certain drawbacks such as poor durability, inadequate cycling performance, low optical properties and low electrical conductivity in its pristine state [124, 128]. These properties thus limit their ability to be used in their pristine state for fiber-based SC and sensors. It is easy to modify the electrical conductivity of CPs by altering the types and amounts of doping. Particularly, the electrochemical performance of CP is improved by the addition of carbon compounds [124]. Numerous electrospinning, hard physical template-guided synthesis (e.g., interfacial polymerization), soft chemical template synthesis (e.g., template-free method, dilute polymerization, reverse emulsion polymerization, etc.), and lithography methods have been used to prepare conducting polymer (CP) nanotubes and nanowires [129]. PANI, PPy, and PEDOT fabrication methods are briefly discussed to understand the specific behavior and working mechanism of conducting polymers and their wide range of applications in chemical, biosensor, and electrochromic display devices, as well as supercapacitors, actuators, and separation membranes.

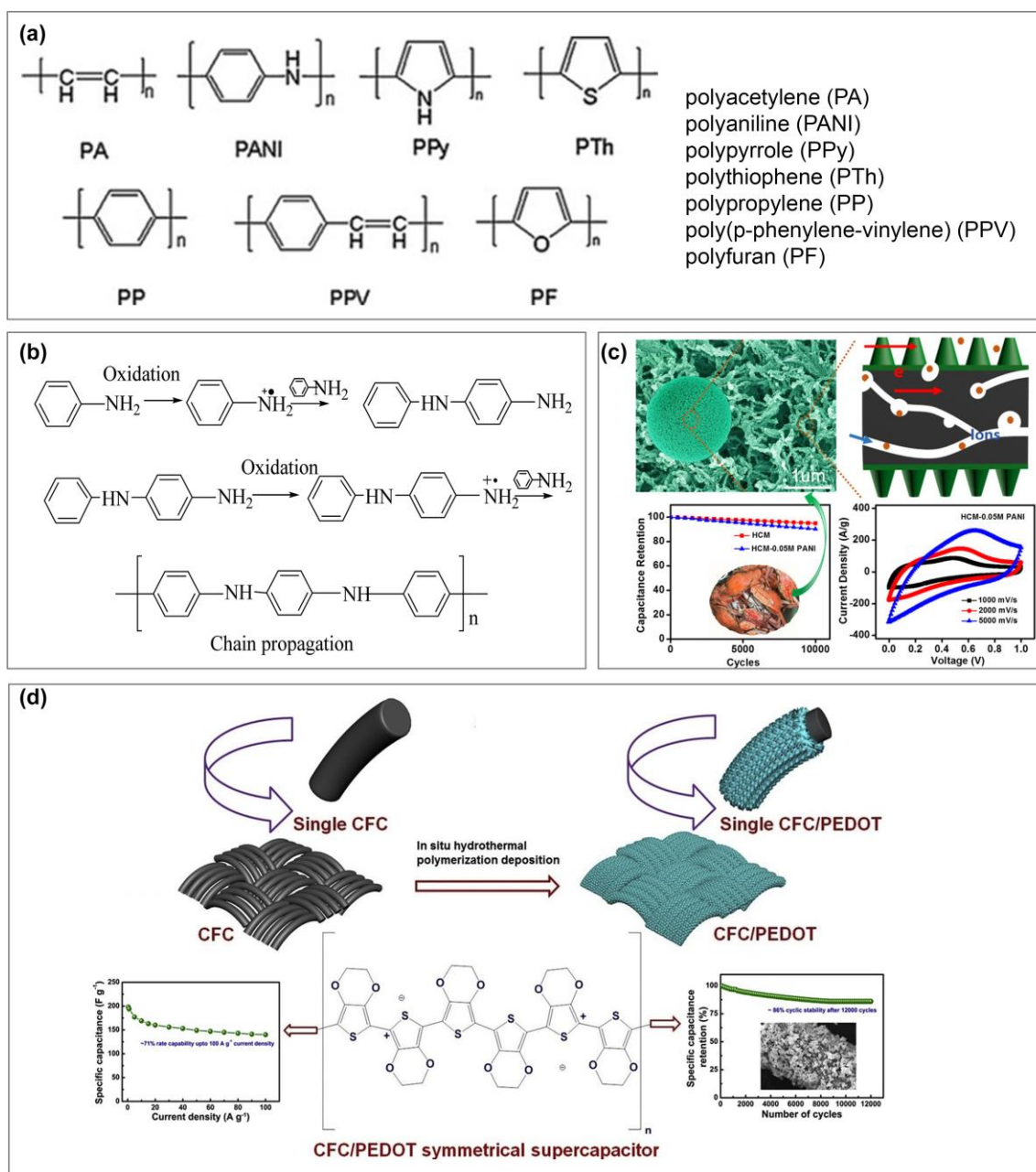


Figure 12. (a) Examples of Conducting polymer's structure. Adapted with permission from Ref. [129]. Copyright 2012 Taylor and Francis. (b) Chemical oxidation of PANi [128]. (This article is licensed under the terms of Creative Commons Attribution 3.0 Unported License). (c) Synthetic and Performance of HCM-PANi. Adapted with permission from Ref. [130]. Copyright 2018 American Chemical Society. (d) Fabrication of SWCNT/PEDOT through in-situ hydrothermal polymerization technique. Adapted with permission from Ref. [131]. Copyright 2017 Elsevier.

### 7.1.1 Polyaniline (PANi)

These are the widely utilised conductive polymers (generated as active materials) in pseudo-capacitance electrodes [132]. PANi has several advantages such as environmental stability, de-doping/base doping/simple acid chemistry, easily synthesized, and tunable conductivity with excellent optical properties [128, 132, 133]. The dopant concentration and  $\text{pH} < 3$  influence the conductivity of PANi fabricated by electrochemical polymerization (interfacial polymerization) and chemical oxidation methods. Under interfacial polymerization, an organic solvent such as an oxidant solution is used to dissolve aniline monomers whereas, under chemical oxidation, an oxidizing agent (like potassium bichromate, ceric sulfate, ammonium persulfate amongst others) is mixed with a monomer precursor of a polymer in the presence of acid [128] (Figure 12b). The electrochemical properties of pristine PANi are generally very poor [132], hence the electrochemical performance can be improved with the introduction of carbon materials [134]. In a recent study [135], flexible PANi-CNF composites were fabricated for SCs using electrospinning and sol-gel techniques for synthesizing CNFs, and in-situ chemical polymerization for coating PANi on the CNFs for better electrochemical performance. Results proved that the PANi-CNFs composite SC exhibited good cycling stability after 1000 cycles (with capacitance retention of about 90%), 234 F/g high capacitance, a 500 W/kg power density with a 32 Wh/kg high energy density. Alternatively, Gao et al [130] reported on hierarchically porous carbon microspheres (HCM)-PANi for symmetric SCs) as shown in Figure 12c. Here, the prepared HCMs were deposited with PANi. The results showed high cycling durability and rate capability after 10000 cycles with 90.6% capacitance retention. Further study reported the fabrication of N-doped carbon microspheres (YC) from yeast via a non-template hydrothermal carbonization route, and PANi was deposited into the hollow carbon microspheres to form YC-PANi hybrids. The electrode SC showed superior cycling stability, a 100  $\text{Fg}^{-1}$  and 500  $\text{Fg}^{-1}$  high specific capacitance at two-electrode and three-electrode systems with a current density of 1  $\text{Ag}^{-1}$  [136].

Aside from the fabrication of SCs, sensors are also developed. In this regard, one-dimensional hierarchical PANi-multi-walled CNTs fibers were fabricated for gas sensing

performance. Here, heating modifications produced n-PANi/CNT and p-PANi/CNT fibers that, for nitrogen dioxide (NO<sub>2</sub>) and ammonia (NH<sub>3</sub>), respectively, showed higher sensitivity and response times [137]. Similarly, PANi/MWCNT composite electrode was synthesized for electrochemical sensing and the modified glassy carbon electrode (GCE) surface of the composite electrode showed excellent phenol detection from the oil field under wastewater [138]. Also, single-walled CN-PANi composite nanofibers were fabricated for chemo sensors, and compared to pure PANi nanofibers, the CN-PANi composite nanofibers demonstrated great sensitivity for gas chemosensors [139].

### ***7.1.3 Poly 3,4-ethylenedioxythiophene (PEDOT)***

One kind of polythiophene (PThs) is PEDOT, with relatively high conductivities compared to other PThs, PANi, and PPy. PEDOT has high electro-optical and electrical properties, high transparency in an oxidized thin film form, good stability and moderate band gap when in an oxidized state [128, 140]. Its insolubility in water is the main problem of PEDOT [128, 141]. Further drawbacks include poor electrochemical stability and low capacitance [142], which have become an important area of research lately to enhance its potential in SCs. In this regard, PEDOT is usually combined with other carbon-based materials to improve its electrochemical stability and enhance its capacitance. For example, a PEDOT symmetrical SC was fabricated where PEDOT nanostructures were deposited on 3D carbon fiber cloth using in-situ hydrothermal polymerization [131] (see Figure 12d). The fabricated SC exhibited a high specific capacitance of 203 F g<sup>-1</sup> at 5 mVs<sup>-1</sup> scan rate with high energy density of 4.4 Wh kg<sup>-1</sup> and minimal degradation at 1000 cycles.

Similarly, PEDOT/CNF composite was fabricated for neurochemical sensing and the microelectrode arrays (MEA) produced electrochemically showed high sensitivity, which proved the effectiveness of PEDOT/CNF composites for neurochemical sensing [143].

### ***7.1.2 Polypyrrole (PPy)***

PPy is also another commonly used CPs because of its enhanced conductivity, high capacitance property, and electrical conductivity, ease of synthesis, high cycling and

environmental stability, good mechanical properties, good thermal stability and low cost. [128, 132, 144, 145]. Electrochemical synthesis like other techniques (Figure 13) for CPs are used for conductive PPy because of ease in the control of the morphology and the thickness.

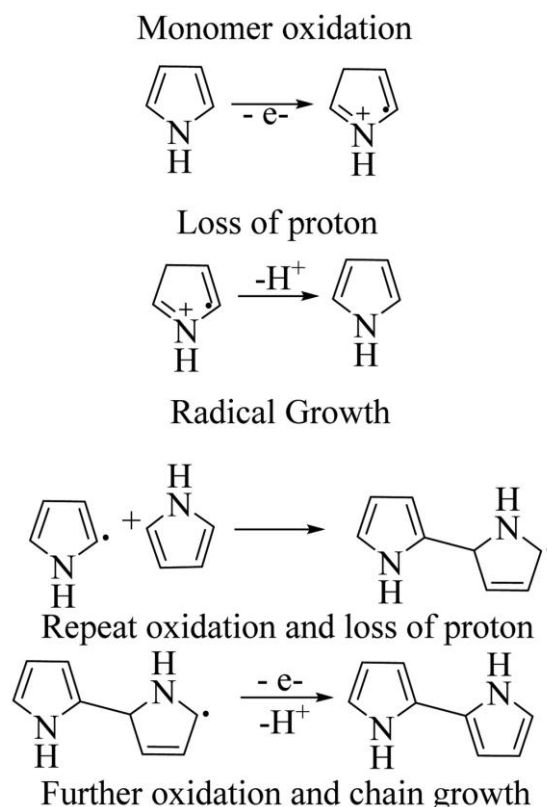


Figure 13. Electrochemical synthesis of PPy [128]. (This article is licensed under the terms of Creative Commons Attribution 3.0 Unported Licence).

In this method, the neutral monomer unit is attacked by free radicals (formed due to deprotonation). The electrochemical performance of PPy is improved using carbon materials, of which most studies have reported on. For example, through electro-polymerization, PPy synthesised on carbon fiber is used to produce a fiber-based SC. The fabricated electrode showed good cycling stability and bending capability, a high specific capacitance of 308.2 F/g [146]. Additionally, CNT-PPy fiber SC was developed using the electrochemical deposition method. The fiber-based SC showed high reliability and stability, 69 Fg<sup>-1</sup> high specific capacitance, 3.6 W h kg<sup>-1</sup> and 3.8 kW kg<sup>-1</sup> high energy density and power density respectively [147]. In another approach, PPy was coated on CNF-MnO<sub>2</sub> via electrospinning, carbonization, and in-situ polymerization. The fabricated CNF-MnO<sub>2</sub>/PPy SC electrode exhibited long

cycling stability, high specific energy of 13.68 Wh/kg, and high specific capacitance of 315.80 Fg<sup>-1</sup>[148].

For drug sensing performance, PPy was electrodeposited on carbon fibers via electro polymerization using cyclic voltammetry in the presence of sodium dodecyl sulfate and potassium nitrate[149, 150]. The synthesized electrode showed good sensing performance for paracetamol.

## **8. Carbon-based composite fibers for SCs and sensors**

Amorphous carbon, graphite, and diamond are the three main types of conventional carbon materials [47]. The properties of carbon vary with the arrangement of the carbon atoms. For instance, the sigma bonding between the diamond's cubic crystal structure and the sp<sup>3</sup> hybridized carbon molecules is what gives the diamond its stiffness and hardness. Weak van der Waals interactions between adjacent layers and strong covalent bonds between sp<sup>2</sup> hybridized carbon atoms in the plane of particular layers characterize the layered structure of graphite. [151, 152]. Several dimensions of new graphitic carbon nanomaterials categorized as two-dimensional (2D) graphene, one-dimensional (1D) carbon nanotubes (CNTs), and dimension-less (0D) fullerene, have been developed recently as a result of advances in nanoscience and nanotechnology. SCs built with carbon nanotubes (CNTs), graphene, and mesoporous carbon electrodes are the most significant energy-storage technologies [47].

Similar to traditional capacitors, electrode materials of EDLCs should have exceptional electrical conductivity and large surface area, attributes that are particularly met by new graphitic carbon nanomaterials. These carbon-based materials are readily available at comparatively low cost, and have good chemical and physical properties, which makes them adapt to a wide range of operation temperatures [153]. Also, these carbon-based materials have tunable properties based on the method of synthesis and can be made into shapes and pore sizes [154].

With the inherent properties of these carbon-based materials, they are widely used as active materials to fabricate carbon-based fibers for SCs and sensors for energy autonomous systems (Figure 14). They have combined properties of mechanical strength, high flexibility,

electrochemical and electrical conductivity [155]. The development of these active materials for electrodes can be derived from synthetic and natural substances for fiber-based SCs in 1D fibers and yarns, 2D and 3D fabrics [156]. The eventual electrochemical properties of a device are influenced by the relevant materials used for electrodes [157].



Figure 14. Overview of source and structure of carbon-based fibers. Adapted with permission from Ref. [155]. Copyright 2020 American Chemical Society.

### 8.1 Materials for SCs and Sensors

Carbon nanotubes, graphene, and activated carbon are the most common materials used in fabricating carbon-based fibers [158]. These materials have inherent properties (Table 5) that enhance their use in various applications.

Table 5. Characteristics of carbon-based materials.

Material	Application	Characteristics	Ref.
Activated carbon	Used as electrodes for supercapacitors.	Ease of fabrication, good electrical performance, require precursors like polymers, bamboo, coal, and starch wood amongst others.	[159, 160]

Carbon nanotubes	Used as energy devices like supercapacitors.	Good electrical conductivity, high chemical and mechanical stability, flexible and durable. Can be split into multi-walled CNTs or single-walled CNTs.	[161]
Graphene	Used as energy storage devices.	Easily accessed by electrolyte, the capacitance of up to 550 F/g, large surface area of $\sim 2620 \text{ m}^2/\text{g}$ , good electronic, cycling capability and thermal properties.	[162, 163]

### 8.1.1 Activated carbon materials

These materials based on their unique properties such as large surface area, pore size, suitable distribution size, good conductivity and chemical stability, they are widely used for SC fabrication [164]. The fabrication of these materials from biomass sources has received attraction due to its abundant nature and low cost [165]. The chemical and physical activation methods used in the production of activated carbons are illustrated in Figure 15a [166]. In a recent study [167], temperature carbon activated (TCA) composite was used to fabricate a mechanically tough high performance SCs. The composite SC exhibited excellent cycle stability,  $5.7 \text{ KWkg}^{-1}$  power density,  $23.7 \text{ Whkg}^{-1}$  energy density and high specific capacitance of .....

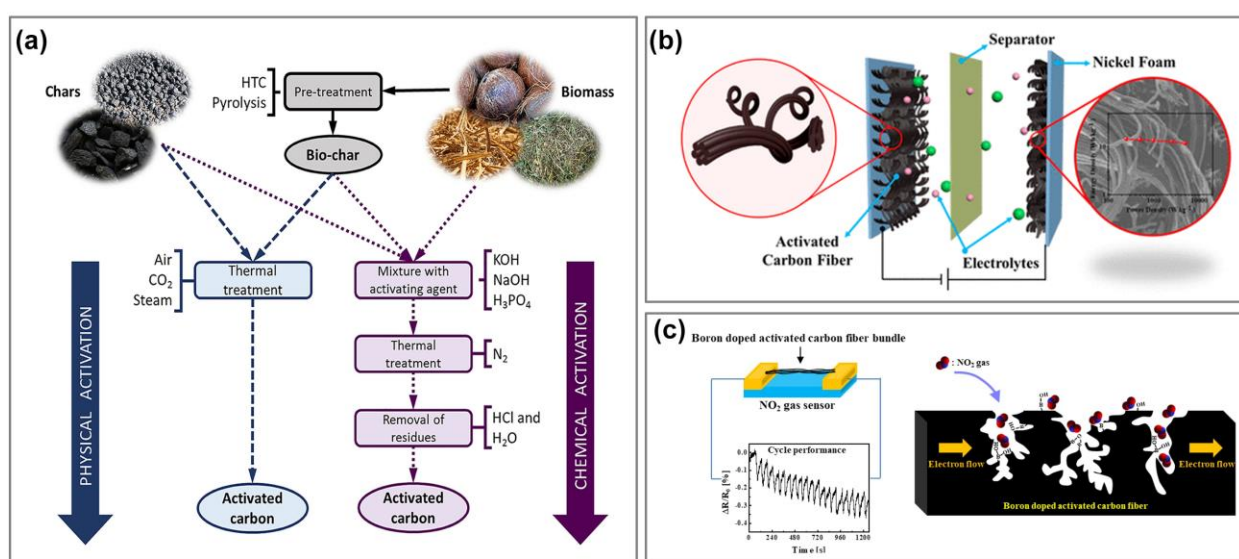


Figure 15. (a) Production flow diagram illustrated chemical or physical activation for

activated carbons [166] (This article is licensed under the terms of the Creative Commons Attribution License - CC BY). (b) Sisal-activated carbon fiber for high performance SC. Adapted with permission from Ref. [168]. Copyright 2019 American Chemical Society. (c) Boron doped ACFs for NO<sub>2</sub> gas sensing. Adapted with permission from Ref. [169]. Copyright 2020 Elsevier.

Extraction of activated carbons (AC) from biomass materials is done through carbonization which is subsequently under an inert atmosphere, activated chemically [170], which shows good power and energy density [171]. In a recent study [172], an activated carbon fiber (ACF) from natural cattail was produced as SC. The activated carbon fiber-based SC showed good electrochemical properties at an activation temperature of 800°C for 2 hours, had excellent cycling stability of more than 97% retention after 10,000 cycles at 1 A g<sup>-1</sup> and had 249 F g<sup>-1</sup> at 0.05 A g<sup>-1</sup> good specific capacity. Similarly, a natural biomass sisal fiber was used to fabricate ACF at different activation temperatures of 700, 750, 800, and 850°C [168].

The potassium hydroxide (KOH) activation method was used to produce the ACF having high specific surface area and good pore size distribution (Figure 15b). At 750°C, the ACF fabricated SC exhibited good cycle stability (93% capacitance retention after long cycle), good capacitance value of 415 F g<sup>-1</sup> at 0.5 A g<sup>-1</sup> and a high energy density of 11.9 Wh kg<sup>-1</sup> in 6 M KOH electrolyte. In another study, waste cotton was utilized to fabricate an activated porous carbon fibers (APCF) for high-performance SC electrode [173]. Three different electrodes (1 M TEABF<sub>4</sub>/AN at 0 - 2.7 V, 0.5 M Na<sub>2</sub>SO<sub>4</sub> at 0 - 1.8 V and 6 M KOH at 0 - 1 V) were fabricated. TEABF<sub>4</sub> at 0 - 2.7 V electrode showed a good long-life cycle, good volumetric capacitances of 74 F cm<sup>-3</sup> at 1 A g<sup>-1</sup>, and good gravimetric capacitance of 112 F g<sup>-1</sup>. The fiber further provided high volumetric and gravimetric energy density. Aside from natural biomass sources, polyacrylonitrile-based precursor fibers were used to fabricate ACF for SC using carbonization and CO<sub>2</sub> activation. Subsequently, through thermal treatments and nitric acids, functioning groups containing oxygen were introduced into ACFs [174]. The fiber exhibited good cycling performance, good pore structures, and a 214 F g<sup>-1</sup> capacitance at a 0.5 mA cm<sup>-2</sup> current density.

ACFs are also fabricated for sensing performance. A recent study [175] synthesized a

single and double-thread ACF for pH sensing. The single thread showed lower hysteresis of 60.4 mV for pH (7-4-7-10-7), and higher sensitivity of 19.66 mV/pH for 2 to 12 compared to the double thread ACFs. These results showed that the fabricated fibers have a good potential for use as a pH sensor. A boron-doped ACFs (Figure 15c) was fabricated for NO<sub>2</sub> gas sensing at normal room temperature [169]. The ACFs contained enriched boron moieties after high-temperature boron doping at 1500°C, which showed rapid, repetitive and selective NO<sub>2</sub> gas sensing. Due to the forming of micropores on the surfaces, the NO<sub>2</sub> gas was easily absorbed and desorbed.

### ***8.1.2 Carbon nanotube composite materials***

CNTs are commonly used as electrodes and latterly as current collectors in the fabrication of fiber-based SCs which come with inherent light weight, mechanical strength and good electrical conductivity [176, 177]. As such, several studies have been conducted using CNTs with different fabrication methods. In a study by Chen et al [178], a coaxial fiber SC was fabricated by synthesising an aligned CNT fiber and a CNT sheet via Chemical Vapor Deposition (CVD). Results showed that the fiber base SC exhibited good power (755.9 W kg<sup>-1</sup>) and energy densities (1.88 Wh kg<sup>-1</sup>), high capacitance discharge of 59 F g<sup>-1</sup>, good electrical conductivity, and tensile strength. These properties influenced the fiber structure in providing an effective area for ion storage and its suitability to be woven into electronic textiles. Similarly, a coaxial fiber SC was fabricated with carbon nanofibers and used as electrodes in coating the outer surface, with the core electrodes coated with MWCNTs (Figure 16a)[177]. The fabricated fiber exhibited excellent cycling performance, high capacitance, surface area, power (13.7 μW cm<sup>-1</sup>), and energy density (0.7 μWh cm<sup>-1</sup>).

To further enhance the electrical conductivity, improve the capacitance and increase the strength of the coaxial SC, a 3D CNT-aerogel electrode fiber SC was fabricated using freeze drying and electrochemical activation (Figures 16b) [179]. The fiber electrode showed long cycling stability, high capacitance (160.8 F g<sup>-1</sup>), good electrical conductivity, high mechanical strength, and a large surface area. In a recent study by Cen and co-workers [180], a novel fiber-shaped asymmetric SC was fabricated using CF/CNT/PPy and CF/CNT/PPy negative

and positive electrodes via an electrochemical method. These were subsequently twisted with PVA-LiCl gel electrolyte to impact good electrochemical properties and a high energy density of  $22.8 \mu\text{Wh cm}^{-1}$ . In furtherance to these studies, composite fiber-based SC was investigated as an energy storage system. In this regard, Yang et al, [181] fabricated an all-solid-state yarn SC using in-situ polymerization of pyrrole and spinning CNT-coated cotton roving. The fabricated yarn SC showed excellent cycling performance, current density ( $0.57 \text{ mA cm}^{-2}$ ) and an area-specific capacitance of  $421 \text{ mF cm}^{-2}$ . Flexible hybrid fibers were fabricated using carbon black(CB)/CNT/manganese oxide ( $\text{MnO}_2$ ). The composite CB/CNT fibers resulted in good electrode capacitance ( $9.2 \text{ F g}^{-1}$ ) and an increased capacitance of  $246 \text{ F g}^{-1}$  with the addition of 0.2 wt% birnessite-type  $\text{MnO}_2$  [182]. Similarly, hybrid nanotube fibers were fabricated for SC using wet spinning. By electrodeposition,  $\text{MnO}_2$  nanoflakes were deposited in the CNTs to form flexible hybrid fibers [183]. These showed stable cyclic retention, a  $14.1 \text{ Wh kg}^{-1}$  energy density, and over  $152 \text{ F g}^{-1}$  specific capacitance.

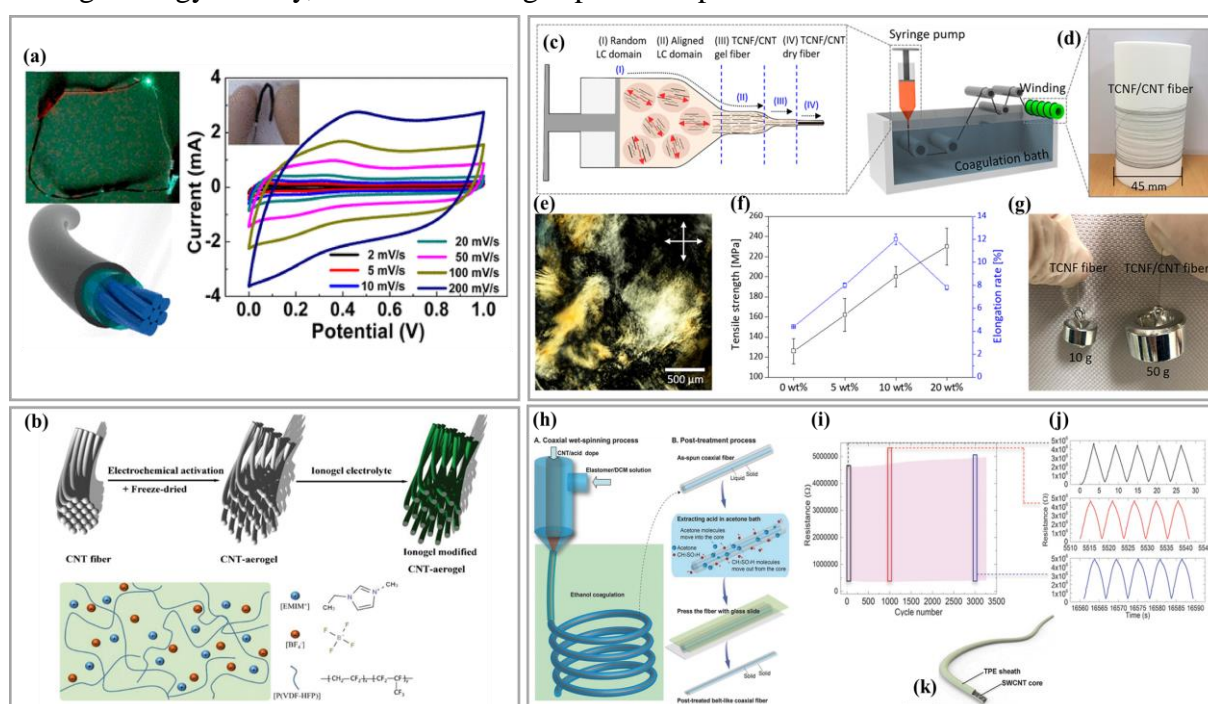


Figure 16. (a) Coaxial fiber supercapacitor from MWNCTs as core electrodes and carbon nanofibers paper as outer material electrodes. Adapted with permission from Ref. [177]. Copyright 2013 American Chemical Society. (b) Fabrication of 3D CNT-aerogel electrode fiber [179]. (c) Fabrication of TCNF/CNT fibers based on hybrid LC spinning technique. (d)TCNF/CNT image collected on the bobbin. (e) Hybrid schlieren LC texture of TCNF/CNT

(20) at 30 mg/mL is shown in a POM picture (f) Mechanical properties of the TCNF/CNT fiber as a function of the CNT content. (g) Photograph of the weights lifted by the single fibers. Adapted with permission from Ref. [184]. Copyright 2019 American Chemical Society. (h) Fabrication processes for the fibers via wet spinning. (i) The fiber's dynamic reaction to stretching and relaxing cycles of 20% to 100% strain at a pace of 400 mm min<sup>-1</sup> demonstrates the sensor's long-term reproducibility after 3250 cycles. (j) Repeatability of the fiber sensor at cycles 1–5, 1000–1005, and 3000–3005. (k) core-sheath structure of the fiber. Adapted with permission from Ref. [185]. Copyright 2018 John Wiley and Sons.

Aside from fabricated CNT-based fibers for SC, other studies have fabricated CNT-based fibers for sensory applications. Poly (lactic acid)/carbon nanotube fibers for glucose biosensors were created in a prior work [186]. The solution-blow spun PLA/MWCNT fibers on indium tin oxide (ITO) electrodes showed a sensitivity of 358 nA mM<sup>-1</sup> and a linear response of 800 mM of glucose. A 3D pressure touch sensor reported by Kim et al, [187], was produced from a network of cellulose/SWCNT fibers. This pressure sensor exhibited high flexibility, a high response speed of <2 ms and high sensitivity of 9.097 kPa<sup>-1</sup>. The fiber could find applications in healthcare monitoring of physiological signals of human motion. With the inherent temperature-dependent CNT fibers, studies have explored these in the fabrication of temperature sensors [188]. Here, a floating catalyst CVD method was used to fabricate CNT fibers which were measured by an *in-situ* technique based on CNT film electric heater [189]. Results showed good temperature sensitivity of -0.15% °C<sup>-1</sup> and an effective reversible electrical resistance response behavior at exposure to heat.

In a study by Cho S-Y et al, [184], they fabricated and reported high-performing tunicate cellulose/carbon nanotube (TCNT/CNT) fibers for sensing abilities (Figure 16c-g). The fabricated fibers can withstand distortions while still maintaining the high sensing performance of NO<sub>2</sub> gas. In a follow-up study, CNTs were wrapped in coaxial thermoplastic elastomers via wet spinning for strain sensing applications as presented in Figure 16h-k [185]. Results showed high linearity and stretchability, high sensitivity of the sensors at 100% strain, and a gauge factor of 425. The fiber-based sensor further showed good repeatability with

cycles of 1-5, 1000-1005 and 3000-3005. In addition, they are used as stretchable interconnects due to the highly stretchable and electrically insulative thermoplastic elastomers.

### ***8.1.3 Graphene materials***

Graphene-based materials have fascinating features such as their highly adjustable surface area, superb electrical conductivity, strong chemical stability, and excellent mechanical performance, make them a good candidate for use in SCs and other energy storage devices [190]. The common techniques for the fabrication of GFs SC are laser reduction technique, hydrothermal strategy, wet spinning, hard template method, electrophoretic self-assembly, and CVD-assisted process. Studies have focused on using graphene with its derivatives to produce fiber-based SC and sensors [191]. To ensure high specific capacitance and electrically conductive fiber electrodes, Zhou et al, [192] developed a hierarchical porous core-sheath graphene-based fiber. The fabricated fiber was achieved by small-sized graphene contained in carbonized phenol formaldehyde resin. The assembled fiber exhibited ultralong cycling life, good flexibility, and ultrahigh specific areal capacitance of  $391.2 \text{ mF cm}^{-2}$  in polyvinyl alcohol/ $\text{H}_2\text{SO}_4$  electrolyte at  $0.1 \text{ mA cm}^{-2}$  in a two-electrode cell. In a similar fashion, Meng and co-workers [193] fabricated 1,4-naphthoquinone (NQ)/rGO on CF for NQrGO/CF negative electrode and nickel-cobalt hydroxide (NCH)/rGO on CF for NCHrGO/CF positive electrode. This graphene fiber-based electrode was assembled into asymmetric solid-state SC (Figure 17a-d). This showed a good cycling performance,  $1642.1 \text{ W kg}^{-1}$  maximum power density and  $50.7 \text{ Wh kg}^{-1}$  maximum energy density. This design for a parallel electrode connection provides advance for fiber-based energy storage. A further study using graphene was conducted where [194] fabricated fiber composites (SBS-G) made of stretchable poly(styrene-butadiene-styrene) and layers of graphene. This fiber composite was modified with carbon black (CB) nanofibers using wet electrospinning. Results revealed the modified CB-nanofibers SBS-G composites had excellent electrochemical properties with a high and improved power and energy densities of  $692 \text{ mW cm}^3$  and  $6.6 \text{ mW h cm}^3$  respectively. The composite fibers further showed excellent cycling stability after 2000 cycles at a 94%

capacitance retention.

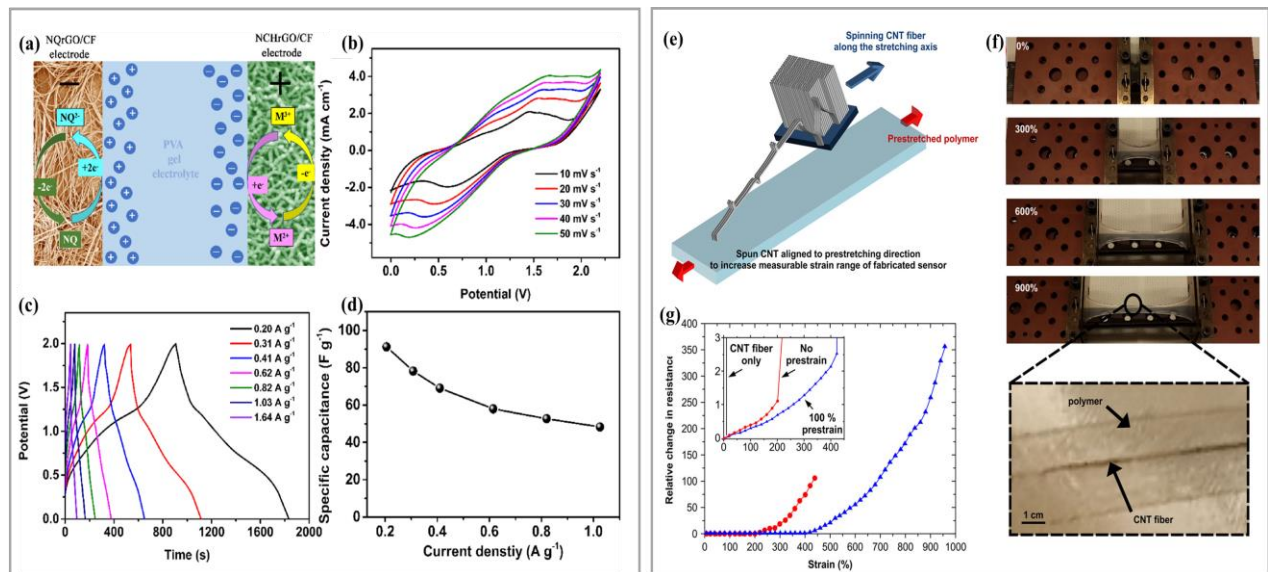


Figure 17. (a) Assembled NChrGO/CF//NQrGO/CF solid-state micro-AFSC. (b) CV curves at various scan rates. (c) GCD curves at various current densities. (d) the plot of capacitance versus the current density. Adapted with permission from Ref. [193] Copyright 2020 Elsevier. (e) Fabrication process for dry-spun CNT fibers attach directly to elastic Ecoflex substrate. (f) Image of strain sensor being stretched parallel to 900%. Graph showing the relative change in resistance as a function of strain for unsupported CNT fibers (black), CNT fibers on an unstrained Ecoflex substrate (red), and CNT fibers on an Ecoflex substrate pre-strained by 100% (blue); the strain varied from 0% to 450% strain (inset). Adapted with permission from Ref. [195]. Copyright 2015 American Chemical Society.

In a different approach, a 3D sheath graphene network was fabricated around a core (graphene fiber) - all-graphene core-sheath fiber (GF@3D-G) as a fiber-electrode [196]. The fiber electrodes exhibited great capacitance behaviour and were lightweight, flexible with high mechanical strength and stability. As such two GF@3D-G electrodes were intertwined to form an all-solid-state fiber SC with H<sub>2</sub>SO<sub>4</sub>-PVA gel polyelectrolyte. Further results showed improved flexibility, fiber shape, stable capacitance of ca. 30-40 $\mu$ F, and good tolerance to repeated straight-to-bending process. To improve the electrochemical performance of rGO fibers, rGO/MnO<sub>x</sub> hybrid fibers were developed by continuous spinning. Here, MnO nanoparticles were well dispersed in NLC rGO dispersions, where the fabrication process is

followed by a chemical reaction. The fibers showed a power density of  $24.76 \text{ mWh cm}^{-3}$  with an energy density of  $2.67 \text{ mWh cm}^{-3}$ .

Sensors made from graphene can sense stimuli like temperature, pressure, light, and chemicals by utilizing the special physical and electrical characteristics of graphene. With their potential for low cost, quick response, and great sensitivity, these sensors have a wide range of applications, from biomedicine to environmental monitoring.

For diverse sensing applications, graphene fiber-based sensors are made using a variety of ways. In this regard, [197] reported the fabrication of a highly selective and ultrasensitive graphene-based single yarn for gas sensing. Using an electrostatic self-assembly, cotton and polyester yarns were coated with reduced graphene oxides (RGOs). The coated yarn sensors exhibited high reliability after several washing and long-term sensing abilities for gas monitoring. It can detect  $\text{NO}_2$  gas at room temperature even at 250 parts per billion. Also, an elastic strain CNT fiber-based sensor was fabricated by dry spinning as shown in Figure 17e-g [195]. This CNT fiber array was directly attached to Ecoflex substrate which provided a good contact area giving off a highly sensitive strain sensing. The fiber demonstrated high durability, responsiveness, and high sensitivity under a stretch of over 900%. Beyond the basic surface coating technique, a highly conductive and stretchable G@PVA fiber was fabricated using a combination of polymer coating and CVD grown ultralong graphene. The fibers exhibited high mechanical stretchability, electrical conductivity and, sensitive response to stretching and bending forces [198]. The fabrication methods emphasize the versatility of graphene in the fabrication of sensors for various applications. Besides the use of pristine reduced graphene for sensor fabrications, functionalized graphene is also used. In a recent work [199], a platinum-functionalised nitrogen-doped RGO fiber (Pt-*n*RGO fiber) on colourless polyimide film was fabricated. The fiber achieved a high sensitivity of 4.52% at 66.4% RH hence making them have a wide range of humidity detection levels. For multi-sensor performance, fabricated copper embedded-RGO which contained semiconducting CuI and Cu metallic particles. Depending on the concentration of Cu, the CRGO fibers demonstrated good chemical sensitivity or temperature insensitivity, and good chemical insensitivity or temperature sensitivity.

## 8.2 Materials preparations for SCs and sensors

The two broad routes for the fabrication of carbon-based fibers, thus the carbonization of precursor fibers and the assembly from nanotubes [155]. The latter can be subdivided into three; solid-phase assembly, liquid-phase assembly and in situ gas-phase assembly from arrays and films, dispersions and aerosols respectively. According to [155], a carbonization temperature of  $>500^{\circ}\text{C}$  is used to process the precursor fibers from natural resources whereas, in the assembly from nanotubes, the fibers from graphene and carbon nanotubes are spun after being synthesized with chemical vapor deposition.

### 8.2.1 Carbonization of precursor fibers

This is a common method used in carbon-based fiber fabrication, where their electrochemical performance, electrical conductivity and microstructure are influenced by the precursors used and the processes undertaken at fabrication [155]. During this process, the polymer undergoes decomposition and rearrangement to form a network of carbon atoms. The resulting material, called carbon fiber, is characterized by its high strength, high stiffness, and low weight, making it useful in a range of applications, including aerospace and sporting goods. The conditions of carbonization, such as temperature and time, are carefully controlled to optimize the properties of the final product. The fiber for carbonization can be achieved in two routes, namely, the pan-based route and the pitch-based route.

(i) Pan-based route: This is the traditional method of producing carbon fibers, where a polymer is heated to a high temperature in an inert atmosphere to form a solid fiber, which is then treated with chemicals to remove the non-carbon elements, leaving a high-purity carbon fiber.

(ii) Pitch-based route: This method involves spinning a precursor material, called pitch, into fibers, followed by carbonization and graphitization to convert the fibers into high-purity carbon fibers. This method is less commonly used but produces fibers with superior mechanical properties compared to the pan-based route.

Natural biomass sources and synthetic polymers are some of the common sources of precursor fibers for carbonization (Table 6). In the carbonization process, the rate and the environment of the precursors' thermal decomposition influence the final carbon material,

hence it is important to control the temperature and the removal of hydrogen, nitrogen and oxygen species [155]. This process produces carbon fibers with the coupling of adjacent carbon chains. It is important that, for carboning pitch or polymer fibers, pre-oxidation is needed before the carbonization process.

Table 6. Common material sources for precursor fibers

<b>Category</b>	<b>Material</b>	<b>Reference</b>
Natural biomass sources	chitosan/chitin	[200]
	wool	[201]
	silk	[202]
	bacterial cellulose foam/film	[203]
	flax, kapok, ramie, cotton, hemp	[204, 205]
	bamboo	[206]
Synthetic polymers	polyethylene, poly(vinyl chloride), poly(vinyl alcohol)-PVA, Polyacrylonitrile-PAN	[207-210]
	mesophase pitch	[211]
	composites or mixtures	

### ***8.2.2 Assembly from nanotubes***

The fabrication of carbon-based fibers by the assembly from nanotubes method involves the production of CNTs and subsequent assembly of CNTs into a fiber. This is commonly accomplished by spinning the CNTs in a solution or generating the CNTs using chemical vapor deposition (CVD). Following their creation, CNTs are aligned and consolidated into a fiber using a variety of techniques, including drawing, twisting, and weaving. The resultant fiber is a composite material that combines the desired mechanical characteristics of the matrix material with the high strength and stiffness of the CNTs. In order to produce high-performance carbon-based fibers with a variety of possible uses, such as aerospace, energy storage, and electronics, this technology is a promising strategy. This process involves the assembly of nano-carbons without complex carbonization processes to form macroscopic carbon-fibers. Such assembly methods utilize either gas or liquid or solid for processing with the required driving forces [155]. This can directly influence the electrochemical, electrical, and mechanical performance coupled with the structure of the carbon-fibers produced.

In-situ gas phase assembly (Figure 19a) breaks down the carbon supply, which is either solid carbon or organic vapor, at high temperatures for carbon atoms, and these develop into CNTs on in-situ produced catalytic nanoparticles floating in the vapor[155]. In the presence of an aerosol, the  $\pi$ - $\pi$  interactions and the van der Waals forces promote the attraction and assembly of CNTs to nano bundles which are subjected to the blowing force of gas. The eventual carbon-based fibers are influenced by condensation treatments, winding speed, and the rate of gas flow. Self-assembly or wet spinning in small tubes are common techniques used in liquid-phase assembly to produce carbon-based fibers. Under self-assembly, an increase in interparticle interactions results in self-aggregation or nanocarbons. The dispersed nanocarbons are extruded under wet spinning as illustrated in Figure 19b into a coagulation bath forming the fibers. Chemical vapor deposition (CVD), scrolling of thin CNTs films, drawing out CNTs from an aligned array with or without twisting and spinning from vertically grown carbon nanotube arrays are common methods used to synthesize graphene (oxide) and CNTs into solid film/array, wet dispersion and aerosol for spinning into fibers [212-216].

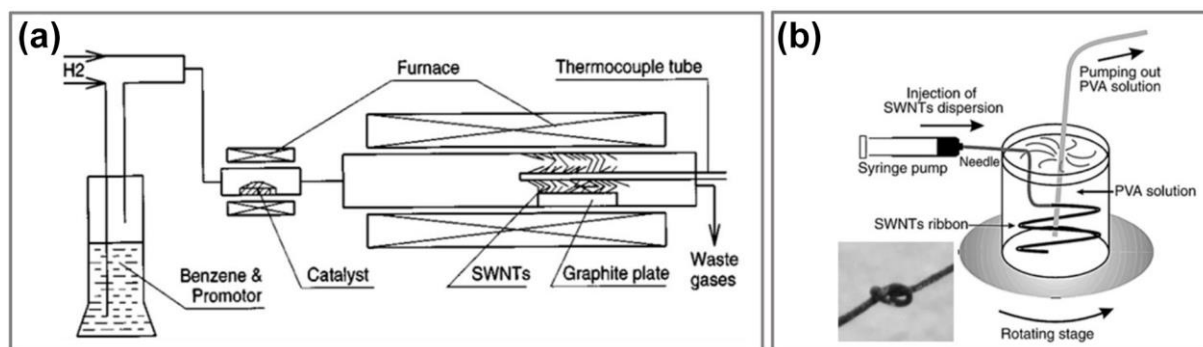


Figure 18. (a) Gas-phase assembly diagram used for floating catalytically decomposing benzene to create SWCNT ropes and ribbons. Adapted with permission from Ref. [217]. Copyright 1998 AIP Publishing. (b) SWCNT fiber is wet spun using revolving coagulant. A fiber knot is shown in the SEM image. Adapted with permission from Ref. [218]. Copyright 2000 The American Association for the Advancement of Science.

## 9. Ethical and Safety concerns

The use of material electrolytes to fabricate fiber-based sensors and SCs has shown excellent properties for their application in wearable sensing and energy storage devices. Aside from these benefits, there are certain ethical and safety concerns regarding the adoption

of these materials for mass customization and production. This largely because scientists fabricate these fiber-based SC and sensors with electrolytes using new techniques are not covered with specific regulations and rules for the manufacturing process [165]. The most important safety concern is the potential flaking-off of harmful active materials, and the leaking and absorption of electrolytes by the human body [219]. Since these occupy a large surface area on the fibers that are in close contact with the human body for sensing performance. For example, the skin absorbs these chemicals which pose serious threats to the organs of the body. The penetration of these nanoparticles via the skin membrane causes abnormalities and death of cells [220, 221]. The long use of these wearable devices without the needed supplementary protective surface could pose serious long-term effects to the human body [222]. This calls for strict regulations and nano safety policies to guide the manufacturing processes and usage by human beings to limit the harmful threats of certain toxic materials. Subsequently, the ultimate disposal of these materials would also pose some serious threats to the environment. With such toxic levels of certain electrolytes used in the fabrication of fiber-based SC and sensors, the burning or indiscriminate disposal would pollute the air, land and possible underground water. Aside from this, the issue of privacy of data extracted from these fiber-based sensors is also another area of concern for consumers. How well data from application areas like health monitoring is secured without sharing is one critical area manufacturers should look at to convince the consumers of maximum protection. There is the need for the development of ethical framework beyond what exist currently that will be based on the principles of safety, environmental responsibility, fairness, transparency, and privacy.

## 10. Future perspectives

For wider commercialization, further research and development is required to maximize their performance, cost-effectiveness, and scalability. Future research development of fiber-based SCs for EAS will focus primarily on the following areas:

- (i) *Development of new materials for electrodes:* In recent years, the development of new materials for electrodes, such as graphene, carbon nanotubes, and metal oxides, has

significantly improved the energy density and stability of fiber-based supercapacitors. It is envisaged that new materials from biomass and their modification will continue. More study of graphene-based nanocomposites, such as those made of graphene/hydroxides or graphene/metal oxides, graphene/conductive polymers, is necessary to meet the long-anticipated need for both high energy and power densities.

New materials, such as MXene, Graphdiyne, nanodiamonds, and transition metal dichalcogenides, are also gaining interest due to their high potential for supercapacitor applications [10, 223-225]. MXene, for example, has a large surface area, outstanding electrical conductivity, and great mechanical characteristics, making it a suitable choice for energy storage. Graphdiyne, on the other hand, has a distinct structure that allows for rapid ion transport and high energy density. Transition Metal Dichalcogenides (TMDs), like other 2D materials, have shown great potential for supercapacitor applications due to their high surface area, good electrical conductivity, and excellent mechanical properties, which are all desirable characteristics for energy storage devices [226]. TMDs also can store energy through both surface and intercalation mechanisms, allowing for high capacitance and energy density. Additionally, TMDs have shown good cycling stability and low resistance, making them promising candidates for use in supercapacitors. However, further research is needed to optimize the performance of TMD-based SCs and to understand their long-term stability.

To enhance the overall Faradic processes across the interface, it is necessary to investigate efforts in the future focusing on the elucidation of nanohybrid structures and the management of the interfacial interaction between graphene and pseudocapacitive materials.

- (ii) *Optimization of fiber properties:* Optimization of fiber properties has enabled the development of high-performance SCs with high energy density and long-term

stability. Further research is necessary electrical and mechanical improvement properties of fibers to enhance the performance of these devices.

- (iii) *Comfort and care for wearable electronics:* Energy storage technologies that are flexible, elastic and malleable are necessary given the increasing growth of flexible electronics. Future research will focus on creating materials based on hybrids that are mechanically flexible, elastic, and fashionable for SCs and other energy storage devices that can be cleaned similarly to normal clothing.
- (iv) *Integration with sensors:* Researchers have explored the integration of sensors with fiber-based SCs to create energy-autonomous systems that can power various applications, such as wearable devices and Internet of Things (IoT) devices; albeit at a laboratory scale. The development of hybrid self-powered or multifunctional commercial systems will be of great interest. Recent ground-breaking research for the integration of flexible SCs with other energy and electrical devices (such as nano-generators, electrochromic devices, Li-ion batteries, and solar cells) has been reported. As a result, integrating carbon-based and other conducting polymers into SCs with these devices will be both beneficial and difficult.
- (v) *Development of scalable manufacturing processes:* A major challenge in the commercialization of fiber-based SCs is the development of scalable manufacturing processes. Researchers are exploring new methods for mass production to reduce the cost of these devices and make them accessible to a wider range of applications.
- (vi) *Advanced analytical tools:* The development of advanced analytical tools, such as computer simulations and characterization techniques, is crucial for understanding the mechanisms of fiber-based SCs and sensors, and for guiding the development of new materials and devices.
- (vii) *Interdisciplinary collaborations:* Collaboration between researchers from different disciplines, such as material science, physics, computer science and textile engineering is necessary to be able to fathom all the nuisances in the development of fiber-based SCs that will satisfy the totality of mankind.

In line with the United Nations Sustainable Development Goals, the development of fiber-based SCs for EAS in the future is currently focusing on the following areas.

- (i) *Carbon-based supercapacitors:* A bright future lies ahead for carbon-based SCs thanks to their high-power density, quick charging and discharging capabilities, and extended cycle life. To make them a competitive alternative to batteries in a variety of applications, including electric cars and the storage of renewable energy, research is now concentrated on increasing their energy density and lowering their cost. To improve their performance, novel materials including carbon nanotubes, graphene, fullerenes, and other carbon allotropes are being investigated. SCs made of carbon appear to have a promising future. It is anticipated that they will be crucial in the shift to a more sustainable energy system. They are also durable and environmentally friendly, making them a sustainable option for energy-autonomous systems.
- (ii) *Cellulose-based sensors:* Sustainable sensors may be produced using fiber-cellulose, which are renewable and biodegradable material. These sensors are economical, environmentally friendly, and simple to include in energy-autonomous systems. Additionally, it is advisable to create and use sensors based on bacterial nanocellulose (BC). BC is suitable for making sensors since it is flexible, highly conductive, and ecologically safe. These sensors can be readily included in energy-autonomous systems and have a high level of sensitivity. for energy-autonomous systems, sensors, and supercapacitors
- (iii) *Chitosan-based supercapacitors:* Chitosan is a biodegradable and renewable material that can be used to create sustainable supercapacitors. These SCs have high energy density and are ideal for energy-autonomous systems that require low-power and long-life energy storage.

The use of sustainable fiber-based materials in the development of SCs and sensors for energy-autonomous systems is crucial for the future. These materials have a high energy density, low environmental impact, and are cost-effective, making them ideal for widespread implementation.

## 11. Conclusion

The high energy and power density, flexibility, and lightweight characteristics of fiber-based SCs and sensors are a testament to the potential applications SCs for energy-autonomous systems. Wearable electronics, Internet of Things (IoT) devices, and portable power systems can all benefit from these gadgets' ability to store and release energy fast. Graphene, carbon nanotubes, conducting polymers, and other materials with various benefits and drawbacks can all be used to create fiber-based SCs. For instance, the large surface area of graphene based SCs results in high energy density, yet their manufacturing costs might be exorbitant. The latter is largely due to the mass-scale difficulty in large-scale synthesis. These can be resolved by ensuring effective reproducibility, uniformity and quality control of graphene flakes. On the other hand, conducting polymer-based SCs has lower energy density but is more cost-effective. Sensors, such as strain and pressure sensors, can also be made from fiber-based materials and integrated into energy-autonomous systems to measure various physical and environmental parameters. These sensors can be used for data collection and analysis, which can help to optimize energy use and extend the lifetime of the system.

The future of fiber-based sensors and SC for EAS remains very promising. There are ongoing efforts for reliability and performance improvement of these devices by fabricating new materials, manufacturing techniques, and device architectures. Fiber-based SCs have the potential to offer high energy density and long cycle life, making them well-suited for use in a wide range of energy-autonomous applications, including electric vehicles. Fiber-based sensors can also be used for varying applications, including structural health monitoring, environmental monitoring and medical diagnostics. These sensors are flexible, lightweight, and can be integrated into a wide range of devices and systems, making them ideal for use in energy-autonomous systems. Largely, there is a growing demand for energy-autonomous systems that are sustainable, reliable, and cost-effective. Fiber-based sensors and SC are poised to contribute significantly to meeting this demand and enabling the development of next-generation energy-autonomous systems.

**Conflicts of interest:** The authors declared no financial/commercial conflicts of interest for this study.

**Data availability:** No data was used for the research described in the article.

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