



Review Article

Wearable Textile Supercapacitors: Material Advancements and Applications

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ABSTRACT

Research into textile supercapacitors (TSCs) for portable and wearable energy storage devices is gaining traction. This is rooted in TSCs' exceptional properties, such as being lightweight and having intrinsic flexibility, stretchability, washability, and compatibility with wearable microelectronic gadgets. This review presents recent developments and advances in the materials and methods used to fabricate TSCs, emphasizing the sustainability of textiles and other commonly used materials in this field. Additionally, we highlight promising environmentally friendly synthetic methods for preparing TSCs. Finally, we offer a perspective on the future directions for further development of TSCs and potential imminent applications of this technology.

1. Introduction

Textile supercapacitors (TSCs) are gaining attention for powering wearable electronic devices. Currently, research and development are focused on TSCs that are greener, more sustainable, and more portable than other mobile energy storage devices such as lithium-ion batteries (LIBs) [1,2]. Although battery technology is developing rapidly, with LIBs dominating the market, they are not the best choice for several reasons. First, LIBs are rigid and bulky and contain hazardous and flammable materials, and their disposal generates significant amounts of waste that are challenging to recycle [3–5]. Second, LIBs' performance degrades after hundreds of cycles of charge and discharge. Lastly, lithium produces a large carbon footprint due to its expensive and resource-intensive purifying processes [6].

A logical and alternative path to power wearables is to incorporate energy harvesting and storage devices into our clothing. This path requires using non-toxic materials and collecting and storing energy from body heat and motion [7]. Integrating electronics into clothing leverages mature textile manufacturing and recycling infrastructures, addressing challenges at the nexus of energy, mobility, and sustainability. This attractive strategy has created much excitement as it utilizes

the long-established social tradition of incorporating textiles into daily life [8]. Textile-based electronics create opportunities for using more sustainable materials than metals and plastics and offer a path to reduce the environmental impact of personal electronics [3,9,10].

The electrochemical performances of supercapacitors depend on the electrode geometry and spacing, specific surface area (SSA), porosity, and electronic properties of the electrode materials. Supercapacitors are among the most promising candidates among various energy storage systems due to their high rate of charging and discharging, high power density, and versatility of design and fabrication methods. Supercapacitors can be manufactured to be light, flexible, and environmentally friendly compared to other energy storage systems. Flexible supercapacitors have been fabricated using materials with high surface area to provide more sites for ion storage and redox reactions during charge and discharge; typically, these materials are based on carbon, metal oxides/hydroxides, and conductive polymers. Carbon materials include activated carbon (AC) particles, carbon nanotubes (CNTs), carbon nanofibers (CNFs), graphene (GN), and carbon fibers (CFs). These components can be used individually or combined in tandem or as composites to achieve improved performance. Although composite electrodes offer a sizeable compositional space of parameters, to date,

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most works studying composite electrodes have been done experimentally, and there is a limited exploration into computer-aided design and modeling of these systems [11–15].

Supercapacitors can be constructed in different ways, as shown in Fig. 1. The first method utilizes the charged electrolyte layer close to the polarized electrodes, called electrochemical double-layer (EDL), to store charge. Capacitors working based on this concept are called electrochemical double-layer capacitors (EDLCs). Firstly, a potential is applied across two symmetrical electrodes separated by an electrolyte, creating net positive and negative electrodes. The polarized electrodes attract opposite charges from the solution, creating a concentration profile close to the electrode. A simplified model accounting for mass transfer coupled with the Poisson equation, assuming the electrodes are non-permeable walls for ions, explains the ion concentration polarization in a charged state. From a classical viewpoint, this zone in solution is divided into Stern and Gouy diffusion layers. The length scale is shown in the inset of Fig. 1a. The charge accumulation on the electrode surface is referred to as the Stern layer. This layer itself is divided into the Inner Helmholtz plane (IHP) and Outer Helmholtz Plane (OHP). The IHP is the locus of the plane passing through the ions that lost their solvation shell and non-selectively adsorbed on the electrode. The OHP is the locus of the plane passing through the centers of the solvated ions next to the adsorbed ions on each electrode's surface. This occurs on the electrodes' surface regardless of the geometry until the curvature of the surfaces reaches the sub-nm range; at this point, the overlapping potential in the cavity leads to selective interactions between charge species and the electrodes, and pore perm selectivity enhances the charge storage [16]. Regardless, in larger cavities of a porous electrode, the classical approach to have the electrode-electrolyte interphase categorized as a

Stern layer with its IHP, OHP, and the Guoy diffuse layer that extends into the quasi-neutral bulk electrolyte nearing the center of pores is widely utilized to explain the observation made upon polarizing the electrodes (Fig. 1d) [17–21]. When the curvature is large enough, the concentration polarization in the diffuse layer plateaus until it reaches electrolyte bulk concentration. Many parameters of electrodes, such as specific surface area, pore structures, and the electrical conductivity of electrode materials, influence the electrochemical properties of capacitors [22]. The electrode materials that create high-performance EDLCs are mostly carbon-based. Materials such as graphene or carbon nanotubes have been among the popular substances researchers utilize in energy storage. EDLCs have a long cycle life but lower energy density compared to the other two methods of charge storage shown in Fig. 1b and c.

The second way supercapacitors are built is through utilizing pseudocapacitance or faradaic response (Fig. 1b). This occurs by applying a voltage difference to two electrodes placed in an electrolyte to create the charge separation as previously described, with one key difference. Once the ions are attracted to the oppositely charged electrode and reach the electrode surface, redox reactions occur between the ions and the electrode [23]. These reversible redox reactions store charge on the electrode to deliver energy when needed. Pseudocapacitors have a higher energy density than EDLCs but a comparatively shorter cycle life because these redox reactions introduce chemical changes to the electrode over time, reducing the number of available redox sites during charge/discharge [24]. The conventional electrode materials used in pseudocapacitance are made of metal oxides/hydroxides and conductive polymers [25]. In recent years, metal carbides/nitrides and sulfides have also been shown to have pseudocapacitive charge storage

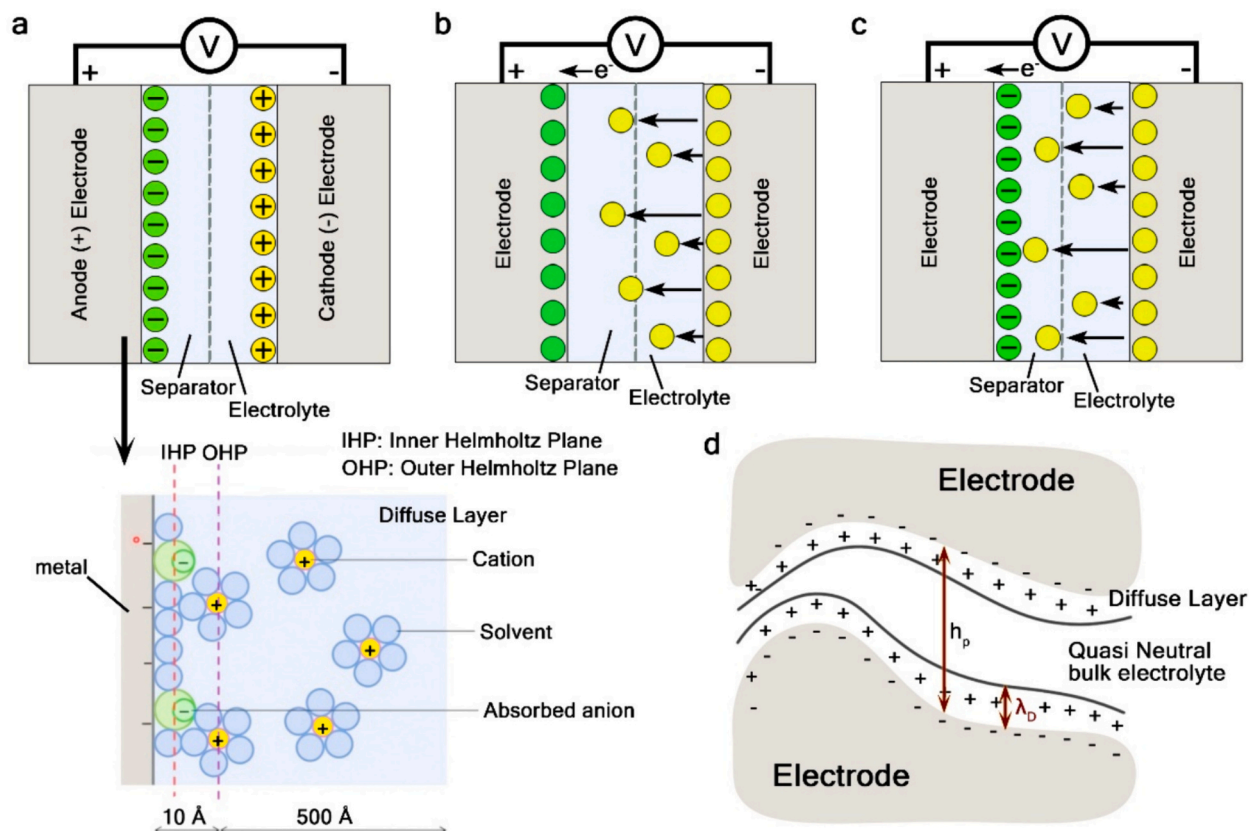


Fig. 1. Schematic illustration of supercapacitor designs: (a) electrochemical double layer capacitor (EDLCs) with inset showing the Stern layer separated into the Inner Helmholtz Plane (IHP) and the Outer Helmholtz Plane (OHP) with ion concentration decreasing in the diffuse layer, (b) pseudocapacitor charge storage behavior where ions are donated into the electrolyte and migrate to across the electrode separator, (c) hybrid capacitive charge storage behavior, and (d) EDLCs in an electrode pore of diameter h_p where the charge separation occurs on the pore wall, within the Debye length (λ_D) and the quasi-neutral bulk electrolyte is in the center of the pore.

properties [16,26–28].

Less commonly used are hybrid capacitors (Fig. 1c), which combine materials that exhibit either EDLCs or pseudocapacitive charge storage behavior to increase the overall specific capacitance that a given supercapacitor can achieve [29–33]. Supercapacitors can be fabricated using asymmetric, symmetric, or hybrid electrodes. Asymmetric supercapacitors contain two electrodes with different materials, while symmetric supercapacitors have two identical electrodes [34–38]. Hybrid electrode systems combine pseudocapacitive and EDLCs energy storage methods and are the preferred type of capacitor for textile-based or flexible supercapacitors [19].

For TSCs to become competitive energy storage devices, high specific capacitance, long cycle life, and rapid charge/discharge are needed. Additionally, the materials should be flexible, stretchable, washable, durable, and non-toxic. Important factors to consider in the design and fabrication of TSCs are the SSA of the conductive materials used, electrode geometry, and electrode spacing. A higher specific surface area provides more sites for EDLCs and redox reactions during charge/discharge and increases the capacitance, while larger electrodes and wider electrode spacing decrease the capacitance of TSCs [39–41]. Three different measurement methods are often employed to find the capacitance of supercapacitors: galvanostatic charging and discharging (GCD), cyclic voltammetry (CV), and electrochemical impedance spectroscopy (EIS). The specific capacitance of supercapacitors is measured in Farads per unit area, volume, or length as determined by the capacitor's configuration. The specific capacitance for stacked or planar/two-dimensional (2D) supercapacitors is reported per unit weight (F/g) or per unit area (F/cm²) [7,42]. The specific capacitance for stacked electrodes can also be measured in per unit volume (F/cm³) [43]. The specific capacitance for one-dimensional (1D) or thread-type supercapacitors is reported in F/cm [11].

TSCs are a rapidly growing research topic in energy storage. Table 1 shows the total number of research articles published during the past ten years related to textile-based supercapacitors extracted from the ISI Web of Science database. These statistics indicate that the number of published articles on textile supercapacitors has increased rapidly since 2015. A review of the studies reported to date shows no report on fully developed commercial TSCs, which means that more work should be conducted to develop wearable TSCs for practical applications. This review discusses the development, sustainability, computational models, process modeling, manufacturing steps, and fabrication methods of TSCs. Practical approaches that can improve the overall performance and address current issues of TSCs are described. Finally, the importance of integrating wearable textile technologies and supercapacitors is highlighted. Special focus is given to life cycle analysis (LCA) and the substrates used for the conductive material.

The main developments in flexible supercapacitors or TSCs are based on the approach taken to fabricate them or the materials used during their fabrication. Initially, TSCs were printed onto fabric using conductive ink, or electrodes were created by coating a fabric with

conductive material where the textile functioned as the electrode separator. One innovation was to create coaxial concentric electrodes that could be stitched into cloth. Still, this approach does not address the resistance that is proportional to the length of the electrodes. Levitt et al. discovered that coating yarn with conductive material and knitting up electrodes drastically reduced the resistance by creating multiple pathways for electrons to travel through the knitted textile [7]. Grube et al. created a new class of TSCs using wool as a substrate for the conductive material in knitted TSCs [44]. Adding conductive material to the dope solution used for spinning fibers was also explored to mitigate delamination of the conductive material from the fiber substrates [7,44,45].

2. Fabrication methods

Textiles used to fabricate TSCs must have several properties, such as flexibility, porosity, stretchability, and mechanical strength [17]. Various methods, such as dip coating, printing, and spinning techniques, can be used to incorporate conductive materials into fabric or yarn to make flexible TSCs [17,46,47]. Flexible supercapacitors can be fabricated in one-dimensional (1D), planar or two-dimensional (2D), and multi-layer or three-dimensional (3D), as shown in Fig. 2 [43,48,49]. Among these configurations, 1D TSCs have concentric electrodes wrapped around a current collector, coated in gel electrolyte, and twisted together to form capacitors, as shown in Fig. 2a. While all these devices have similar potential applications and can be incorporated into clothing, each construction method has its advantages. For instance, 3D and 2D flexible supercapacitors have many potential applications in personal electronics, wearable devices, biomonitoring, and smart textiles [50], while one-dimensional fiber or yarn-based supercapacitors (YSCs) are suitable for implantable medical devices or stitching electrodes into cloth due to their smaller size and greater flexibility [51].

2.1. One-dimensional (1D) fiber/yarn supercapacitors

In 1D fiber supercapacitors, the electrodes, electrolyte, and current collectors are all combined into 1D fiber-shaped supercapacitors and wrapped together to form a single yarn. This type of fabrication [52] reduces the size of the flexible supercapacitors, as shown in Fig. 2a. Yarn supercapacitors (YSCs) can be prepared by spinning graphene, CNTs, or metal oxides into continuous fibers to form conductive filaments woven or stitched into cloth, increasing their versatility in practical applications [53–55]. These conductive fibers can also be wet spun into conductive yarns/filaments to improve their mechanical strength and flexibility [56,57]. Combining these conductive fibers with non-conductive substrates decreases the concentration of conductive material in YSCs, an important factor in defining their capacitance [58]. YSCs are usually characterized by twisting two yarn electrodes with a gel separator before connecting to a potentiostat to collect CV, EIS, and GCD data, as shown in Fig. 2a [49].

YSCs can be used for medical implants to provide continuous power to these devices or stitching supercapacitors into cloth [49,51]. Along these lines, Shi et al. [59] studied coaxial wet-spun yarn supercapacitors and showed high areal capacitance (up to 214 mF cm⁻² at a scan rate of 5 mV/s), energy density (42.8 μWh cm⁻²), and excellent cycling stability of 83.58 % capacity retention after 5000 cycles. Shi's group also reported that the capacitance and energy of coaxial fiber-shaped supercapacitors with concentric electrodes also increased proportionally with the increased device lengths. Peng's group reported the specific capacitance of coaxial supercapacitors is tenfold higher than that of twisted ones [60].

As previously mentioned, wet spinning and electrospinning techniques are also employed to produce textile supercapacitors. The wet spinning method creates fibers of polymeric materials combined with carbon or metals to provide both EDLCs and faradaic or pseudocapacitive responses. Wet spinning techniques can create fibers with small diameters and high surface areas, which can be flexible, lightweight, and

Table 1
Total annual publications in the field of textile-based supercapacitor (including fibers, yarns, and fabrics).

Year of publication	Number of publications
2014	56
2015	67
2016	134
2017	183
2018	198
2019	181
2020	194
2021	201
2022	174
2023	168
2024	77

Data source: ISI web of knowledge, Accessed on 07/04/2024.

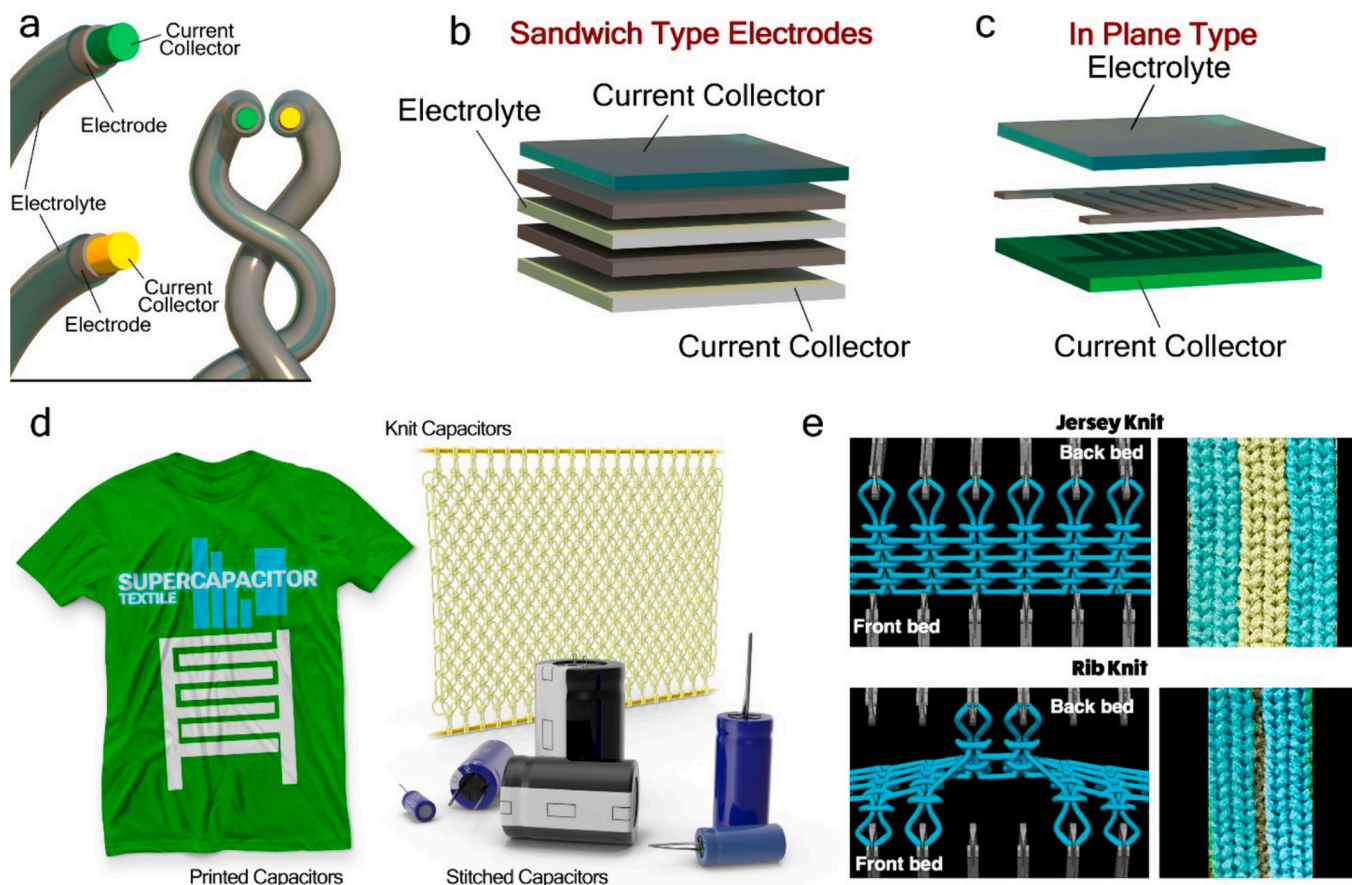


Fig. 2. Different supercapacitor configurations for integration into wearables: a) thread-type supercapacitors with electrodes forming a shell around current collectors with a gel electrolyte separator before twisting around each other; b) sandwich-type supercapacitors where a gel electrolyte layer is sandwiched between two electrodes which are in turn sandwiched between current collectors; c) in place type supercapacitor printed onto a current collector with a layer of electrolyte on top; d) different configurations for incorporating flexible electrodes into existing fabric structures; e) comparing the types of machine knit stitches and their effect on electrode size and spacing. Reproduced with permission [7].

elastic [61–63]. The electrospinning technique uses electrical energy to draw threads of various solutions into fibers. The fibers produced by this technique can be controlled in terms of morphology, porosity, and composition to achieve high electrochemical performance [64,65].

Potential drawbacks of 1D fabrication methods of YSCs, are the linear relationship between resistance and YSC length, the lower capacity, and the increased risk of short circuits caused by an uneven coating on YSCs [66,67]. Some address the issue of uneven coating by creating core-sheath threads of concentric electrodes with a conductive center and a fiber or polyelectrolyte-wrapped exterior; however, this approach decreases the quantity of effective material and does not address the proportional relationship between resistance and length [60,68,69]. The coaxial structure can improve the performance of fiber electrodes and address common problems in YSCs by decreasing the electrode spacing while eliminating short circuits if the YSCs are brought into contact or entangling themselves [59,69].

2.2. Two- and three-dimensional flexible supercapacitors

Multi-layer 3D supercapacitors are constructed by placing two electrodes on either side of an electrode separator and submerging the supercapacitor in an aqueous electrolyte, as shown in Fig. 2b; alternatively, a gel electrolyte can act as a separator and eliminate the need for the liquid electrolyte [70,71]. This method requires the separate preparation of both electrodes before the TSCs' assembly. To date, very few multi-layer SCs have been introduced, and research has focused more on single-layer textile-based supercapacitors, as shown in Fig. 2c. Usually,

the conductive material is loaded onto woven cloth or individual yarns, stitched, or knit into TSC formation, as shown in Fig. 2d. Fig. 2e shows how the different knitting stitches available for machine knitting TSCs, effect the electrode size (teal) and spacing (yellow).

One strategy for preparing conductive cloth or yarn is dip coating. This method is cost-effective, easy to use, and scalable, and it has been the most common approach to loading conductive materials onto yarns. This method involves saturating fabrics or yarns in a conductive solution [29,72]. An alternative to dip coating is spray coating, which allows for depositing thin layers of materials on flexible substrates [73]. Another approach is drop casting, in which conductive ink is applied to a substrate, and as the solution evaporates, a conductive film forms. The disadvantage of this technique is the lack of control over the thickness of the material and the difficulty in obtaining uniformity [19,74]. Bar coating deposits ink on a substrate and uses a bar to remove the excess solution to make a uniform coating. This method can produce thin-layer substrates with a large surface area [75]. A commonly shared disadvantage of these methods is the stiffening of the fabric upon processing. This is due to the need for sequential coatings of the ink to achieve the desired conductivity [19]. Another issue that needs to be addressed is the degradation of the coating when it is worn or washed, causing the conductive coating to delaminate from the substrate. Several researchers have used electrolytes, polymers, binders, and resins to encapsulate or protect the ink so the conductive coatings do not deteriorate [72,76].

Textile supercapacitors can also be fabricated by inkjet and screen printing. These printing methods are advantageous because they allow conductive materials or ink to be deposited in specific patterns on a

substrate and control the amount of ink or materials used to produce TSCs. Both methods are fast, easy, scalable, and can create flexible electrodes, but each technique has its advantages and disadvantages [77]. Due to its high precision, inkjet printing can achieve complex designs. However, it requires small particle size and low ink viscosity to avoid clogging the machines used for this technique [37,77]. The screen-printing technique is used to deposit conductive ink on a substrate through a mesh screen, reducing material waste. This method has a high deposition rate and allows for excellent flexibility and stretchability of the printed systems without sacrificing capacity or mechanical strength [8,78–80]. Screen printing is unsuitable for delicate designs, and the prints' quality depends on mesh structure and ink viscosity. There are also issues with the coating uniformity and stability, and defects due to the scratches and creasing of the print are common [19,81].

Single-layer/2D supercapacitors have sandwich or planar structures, and their fabrication requires fewer steps than those needed to make 3D supercapacitors, as shown in Fig. 2b. Instead of preparing multiple layers separately, single-layer capacitors are usually made by applying a conductive coating to fabric to print circuits [82]. This approach suffers from a high risk of delamination for the conductive coating applied on the flexible substrate, primarily upon bending or movement [83]. Conductive coatings can be applied to individual yarns before knitting into conductive clothes or circuits [7]. This innovative method of coating individual fibers and single yarns before textile manufacturing decreases resistance by creating more contact points within the yarn and throughout the knitted fabric through which electrons can flow [84]. Conductive ink can also be used to print circuits onto fabric or apply films to either side of a textile base that acts as the electrode separator before introducing an electrolyte [82,85]. These methods further reduce the number of steps required for fabricating supercapacitors.

There are many examples of 2D and 3D TSCs designed for applications in biomonitoring for sports and medicine, with a few examples summarized here. Levitt et al. created a planar electrode out of $\text{Ti}_3\text{C}_2\text{T}_x$ dip-coated cotton yarn knit into a capacitor with a polyester separator soaked in polyvinyl alcohol (PVA)/phosphoric acid electrolyte and achieved a specific areal capacitance of 707 mF/cm^2 with a 100 % capacitance retention after 10,000 cycles [7]. Zhou et al. created a sandwich-structured supercapacitor by printing squirrel-shaped activated carbon ink electrodes on either side of a piece of cotton cloth soaked in PVA/phosphoric acid electrolyte to form the electrode separator and support for the conductive material; their system achieved a 63.7 F/g specific gravimetric capacitance with an 85 % capacitance retention after 2000 charge/discharge cycles and an 89.7 % capacitance retention after 100 bending cycles, and a 93.3 % capacitance retention after 100 twisting cycles [28].

3. Electrode materials for textile supercapacitors (TSCs)

Textile supercapacitors are typically fabricated by depositing electroactive materials, such as MXenes, conductive polymers, metal oxides, nitrides, or sulfides, onto flexible fabric or yarn substrates [12,13,19]. Desirable materials for TSC electrodes must be non-toxic and have a large specific surface area, high conductivity, adequate mechanical, chemical, and thermal stability, acceptable corrosion resistance, and low cost. The materials determine the energy storage mechanism; for instance, some oxides, carbides, nitrides, sulfides, and conducting polymers present pseudocapacitance, and most carbonaceous materials demonstrate EDLCs. Thus, hybrid capacitors can be constructed by leveraging both mechanisms in the TSC design [52,86].

3.1. Carbonaceous materials

Carbon materials such as activated carbon, carbon fibers, CNTs, and graphene are popular for creating electrodes for wearable textile supercapacitors. These materials provide high electrical conductivity, a large surface area, diverse compatibility with composite materials, and

acceptable chemical and electrochemical stability. For porous electrodes made of carbonaceous materials, the pore structure, shape, size distribution, and surface functionalities control the performance of textile supercapacitors [87,88].

Activated carbons like carbon black are a group of materials characterized by highly porous, almost spherical carbon particles of colloidal size, which can be produced by thermal decomposition or partial combustion of hydrocarbons in the gas phase. Activated carbon is preferred as an electrode material compared to other carbon-based materials due to its low cost and environmentally friendly nature. Increasing porosity increases the SSA, providing more sites for EDLCs or redox reactions when activated carbon is used as an electrode material [89,90]. Activated carbon has been derived from many materials, such as silk, wool, coal, coconut shells, and wood. Many commercial supercapacitors use activated carbon obtained from coconut shells because they have more micropores than activated carbon made from charcoal. Activated carbon synthesized from silk and wool is naturally doped with sulfur and nitrogen because these fibers naturally contain those elements [91–94]. Food waste and other biowastes produced by various industries have also been explored as raw materials for creating activated carbon [91,94]. Taking this pathway reduces waste produced by many industries [95].

Activated carbon fibers (ACFs) are porous carbons in a fiber-like structure and are considered a hybrid form of activated carbon and carbon fibers. ACFs can be prepared via thermal treatment of carbon fibers. The advantage of an AFC electrode is its low electrical resistance and potential for good contact with the current collector. These electrodes mainly possess double-layer capacitance, with some pseudocapacitance attributed to their micropores [96,97]. Zhai et al., 2015 [98] fabricated yarn supercapacitors from activated carbon and carbon fibers and exhibited a specific capacitance of 45.2 mF/cm^2 at 2 mV/s , a power density of 27.5 μW/cm^2 , and an energy density of 6.5 μWh/cm^2 .

Carbon nanotubes (CNTs) are alternative materials for creating conductive electrodes. They can be fabricated as single-walled carbon nanotubes (SWCNTs) or multi-walled carbon nanotubes (MWCNTs) and are composed of an arrangement of hybridized carbon atoms. CNTs can be deposited on carbon fibers or spun into yarn [11,99]. SWCNTs are formed by rolling up a single sheet of graphene, while MWCNTs are formed by rolling up multiple graphene sheets [100]. The rolled configuration of CNTs creates high electrolyte accessibility, reduces the length of charge transport routes, and results in more rapid charge transfer and higher conductivity. CNTs also have high mechanical strength and flexibility, both valuable properties in textiles, making them a popular choice for textile electrode fabrication [101].

Carbon nanotubes are produced using chemical vapor deposition (CVD), laser ablation, and carbon arc discharge. The arc-discharge method uses high temperatures (around 4000°C) to evaporate graphite electrodes with electrical charges, but this method produces CNTs with high impurity levels. Laser-vaporization technique (LVT) uses high-power lasers with high-temperature furnaces to grow CNTs from high-purity graphite. LVT produces high-purity and high-quality CNTs, but the low production rate restricts scalability. CVD uses catalyst-assisted thermal decomposition of hydrocarbons to synthesize CNTs. This method is the most popular technique for synthesizing CNTs due to its high yield and efficiency [102]. CNTs have been used in textile supercapacitors due to their extraordinary properties, such as high specific surface area, porosity, higher interconnection of active materials, unparalleled electrical conductivity, superb mechanical robustness, chemical and thermal strength, and low mass density [103]. The critical factors that influence the capacitance of electrodes are the morphology, number of walls, surface area, size distribution, structure, and purity of CNT electrodes. Patel et al. [86] fabricated a fiber-shaped supercapacitor by wrapping aligned CNT sheets on an elastic fiber with high stretchability and specific capacitance (19.2 F/g at 0.1 A/g).

Graphene is another well-known candidate for preparing electrodes in electrochemical systems and textile-based supercapacitors. This is due

to its outstanding properties, such as high electrical conductivity, high carrier mobility, large specific surface area, strong mechanical strength, and outstanding chemical and thermal stability. Graphene has a high carbon content and is arranged in a 2D crystal honeycomb structure, providing a high SSA, as high as $2675 \text{ m}^2/\text{g}$ [104–106]. Graphene's large surface area and hydrophilicity make it desirable for TSC research, especially when combined with other materials to create composite electrodes [107]. Graphene achieves superior supercapacitor performance compared to carbon-based materials such as AC and CNT [106]. Graphene sheets are synthesized either by CVD or mechanical, thermal, and liquid phase exfoliation. Graphene oxide (GO) is a graphene derivative and can be obtained by treating graphite material with strong oxidizing agents. The basic properties of GO are large surface area, strong mechanical strengths, appealing electronic and optical properties, high stability, and layered structure [52,108]. Reduced GO (rGO) is another graphene derivative and exhibits properties of both graphene and GO [109,110]. Graphene can be combined with CNTs to create conductive yarns by wet spinning coaxial electrodes [69]. Graphene has also been combined with polymers to create stacked electrodes on top of textiles [43]. Table 2 reviews the electrochemical performances of textile supercapacitors based on carbon materials.

3.2. Conductive polymers

Conductive polymers are pseudocapacitive materials and are another popular option for preparing electrodes for wearable and flexible supercapacitors due to their reversible redox nature, high charge density, excellent conductivity, affordability, biocompatibility, acceptable interactions with textiles, and low-temperature synthetic process. Conductive polymers can conduct electricity through a conjugated bond system along the polymer chains. As shown in Fig. 3a, the (π) electrons are delocalized along the polymer chain. As a result, different energy levels within electronic bandgaps of conjugated polymers are accessible. The delocalization of the electrons defines the state of charge of conjugated polymers, manifested by their degree of oxidation, loosely referred to as doping. In most cases, the dopants are negative ions, and the doped polymers are often p-doped. There are reports on n-doping of conjugated polymers, but for most cases, p-doped materials are more commonly researched. Conducting polymers can be doped electrochemically, and the change in polymer between its reduced and oxidized forms can be utilized to create pseudocapacitors. The repeated electrochemical charge/discharge is concomitant with swelling and shrinking as the solvated ions are pulled in and out of polymers under potential. This repeated volume change results in a decreased charge/discharge cycle lifespan. In doped states, polymers are considered charged and conduct electricity, and when discharged, they act as insulators [143].

Polyaniline (PANI) (Fig. 3b), polypyrrole (PPy) (Fig. 3d), polythiophene (PT) (Fig. 3c), and their derivatives have been commonly investigated as active electrode materials for textile supercapacitors. These polymers can be used as conductive coatings over less conductive substrates, such as metal oxides, to enhance their conductivity and energy density required to improve device capacitance. Electrodes made solely from these conductive polymers can be prepared by inkjet printing or drop-casting of their solutions onto a substrate [144–146].

PANI has attracted much interest in supercapacitors because PANI electrodes provide multiple redox reactions, excellent flexibility, ease of fabrication, and low cost [52]. PANI is a conductive polymer with a conjugated single and double pi-bond system that gives this conductive polymer its ability to transport electrons along its backbone; however, the specific capacitance of PANI is highly dependent on the synthetic conditions, and the poor cycling stability of PANI has limited its application in supercapacitors [147]. In addition, PANI is typically synthesized through oxidative polymerization in an aniline bath, leaving behind significant toxic waste. Currently, greener methods with lower environmental impact are being pursued to process PANI, making it a more attractive choice to be combined with other active materials to

fabricate electrodes [144,148,149]. PANI has three states, fully reduced leucoemeraldine, partially reduced emeraldine-the most conductive, and fully oxidized pernigraniline, all shown in Fig. 3b, where the X subscript indicates the degree of oxidation, and n indicates the number of repeat monomer units in a polymer chain. During reduction, p-type doping of PANI to form leucoemeraldine forms electron holes, causing electrons along the backbone to move into neighboring holes and switching the locations of the single and double bonds, resulting in the charge transport along the length of the conductive polymer [150,151].

Similarly, polypyrrole is a conductive polymer commonly used for flexible and wearable supercapacitors because of its ease of fabrication, good redox properties, acceptable electrical conductivity, biocompatibility, non-toxicity, sustainability, and adequate thermal stability [152,153]. The disadvantages of PPy are its brittleness, difficulty in doping/de-doping, high cost, and poor cyclic stability. However, PPy can be combined with other fiber polymers or nanocomposites to overcome the noted challenges and drawbacks [154]. PPy has a similar charge transfer process to PANI, and can be doped electrochemically to facilitate adsorption and desorption of ions into the conductive polymer [151,152,155]. The neutral, oxidized, and reduced forms of PPy are shown in Fig. 3d.

Polythiophene and its derivatives are conductive polymers with interesting properties, such as excellent conductivity in its oxidized (doped) state, flexibility, low cost, pseudocapacitance, processability, and electrical properties. Fig. 3c shows the base and p-doped PT. The disadvantages of polythiophene are low cycling stability, low specific capacitance, and low conductivity in its neutral form [52]. To address these issues, polythiophene derivatives are compounded with other active materials such as polystyrene sulfonate, metal oxide, or conductive nanocomposites to increase conductivity and enhance specific capacitance [146,156,157]. Table 3 summarizes the electrochemical performances of textile supercapacitors based on conductive polymers to date.

3.3. Metal oxides, carbides, nitrides, and sulfides

Metal oxides are the most common materials used in flexible and wearable textile supercapacitors due to their wide variety of oxidation states required for redox charge transfer at the electrode surface [21,173]. Metal oxides are typically produced through hydrothermal, solvothermal, and wet-etching techniques. For instance, metal oxide can be prepared by heating a metallic precursor in water or solvents. Hydrothermal synthesis methods are considered the most robust and efficient approach that leads to highly pure products with high levels of crystallinity [174]. Microwave-assisted heating is another method for producing metal oxides. It is faster and consumes less energy than hydrothermal synthesis [175].

Manganese dioxide (MnO_2) is the most attractive metal oxide for textile supercapacitors due to its low cost, nontoxicity, and environmental friendliness [52]; however, it has poor cycling ability [52,176]. To address these issues, MnO_2 is usually combined with highly conductive materials such as CNTs and conductive polymers for use in TSCs [177–179]. Nickel oxide (NiO) is also used in TSCs due to outstanding properties such as low cost, ease of preparation, non-toxicity, and environmental friendliness. The poor electrical conductivity and low SSA of NiO -based electrodes can be improved by forming NiO as nanostructures. This leads to an increased number of active sites that boost pseudocapacitance [180–182]. Cobalt oxide and hydroxide are common materials used in supercapacitors because of their outstanding reversible redox reactions, high surface area, cycle stability, and excellent corrosion stability [183]. Titanium oxide has also been used in supercapacitor research because of its high sustainability, ease of synthesis, high energy density, non-toxicity, and high purity [175].

In the past decade, transition metal carbides/nitrides (i.e., MXenes) have attracted tremendous research interest in the field of textile supercapacitors due to their extraordinary properties, including large

Table 2

Summary of the electrochemical performance of various flexible textile-based supercapacitors based on carbon materials.

Electrode material	Flexible substrate	Fabrication Process	Electrolyte type	Electrolyte material	Capacitance	Energy density	Power density	Cycling stability		Ref.
								Cycles	Capacitance retention (%)	
Activated carbon and carbon fibers	Carbon fiber yarns	Chemical deposition	Polymer gel	PVA/H ₃ PO ₄	45.2 mF/cm at 2 m V/s	6.5 μ Wh/cm	27.5 μ W/cm	10,000	86.6 %	[98]
Activated carbon/cotton	Cotton lawn and polyester microfiber	Screen printing technique and dip coating	Aqueous	1 M Na ₂ SO ₄ and 2 M Li ₂ SO ₄	430 mF/cm ² at 5 mA/cm ² or 85–95 F/g at 1–10 mV/s	N.R.	N.R.	10,000	92 %	[111]
Activated carbon cloth electrode	Carbon cloth	Thermal annealing	Polymer gel	PVA/H ₂ SO ₄ (electrode)	88 mF/cm ² (8.8 mF/g) at 10 mV/s	N.R.	N.R.	20,000	95 %	[112]
Activated carbon@carbon fabric	Carbon fiber	Screen printing technique	Solid polymer electrolyte	PVA/H ₂ SO ₄ (SC devices)	15.3 mF/cm ² (0.765 mF/g)	N.R.	N.R.	200	~80 %	[8]
N-doped carbonized cotton	Cotton	Chemical vapor deposition system	Aqueous	PVA/H ₃ PO ₄ /silicotungstic acid	0.51 F/cm ² at 10 mV/s	N.R.	N.R.	200	~80 %	[8]
N-doped carbonized cotton	Cotton	Chemical vapor deposition system	Aqueous	1 M H ₂ SO ₄	207 F/g at 1.0 A/g	7.2 Wh/kg	3823 W/kg	10,000	134 %	[113]
Carbonized bamboo fibers	Bamboo fiber	Carbonization of bamboo fiber	Aqueous	KOH	512 F/g at 0.4 A/g	54 Wh/kg	7.9 kW/kg	5000	~100 %	[114]
Nanoporous carbon from corn silk fibers.	Corn silk fiber	Hydrothermal carbonization process	Aqueous	6 M KOH	~160 F/g at 1 A/g	~32.28 Wh/kg	870.68 W/kg	2500	~ 92.5 %	[115]
SWNT/cotton	Cotton fiber	Dipping and drying process	Aqueous	2 M Li ₂ SO ₄	~70–80 F/g at 0.1 A/g	N.R.	N.R.	35,000	99 %	[116]
MWCNTs/coated fabric	Cotton fabrics	dip-pad-dry process	Polymer gel	(PVA)/H ₃ PO ₄	8.24 F/g (9.18 F/cm ²) at 5 mV/s	6.30 Wh/kg	1.12 kW/kg	5000	96.3 %	[103]
MWCNTs/cotton	Cotton fabrics	Screen printing method	Polymer gel	PVA/KOH	26.4 F/g (13.8 mF/cm ²) at 10 mV/s	N.R.	N.R.	2000	90 %	[117]
A carbon microfiber bundle coated with MWCNTs	Carbon microfiber Bundle	spray-coating	Polymer gel	PVA-H ₃ PO ₄	6.3 mF/cm (86.8 mF/cm ²) at 2 mV/s	0.7 μ Wh/cm	13.7 μ W/cm	1000	94 %	[118]
						9.8 μ Wh/cm ²	189.4 μ W/cm ²			
Electrode material	Flexible substrate	Fabrication process	Electrolyte type	Electrolyte material	Capacitance	Energy density	Power density	Cycling stability		Ref.
								Cycles	Capacitance retention (%)	
CNT sheets coated on elastic fiber	Elastic fiber	Chemical vapor deposition	Polymer gel	PVA-H ₃ PO ₄	41.4 F/g at 0.1 A/g	0.363 Wh/kg	421 W/kg	1000	90 %	[45]
(CNTs)/CNF/CF	Carbon fabric	chemical vapor deposition	Aqueous	0.5 m Na ₂ SO ₄	140 F/g at 5 mV/s	N.R.	N.R.	2000	95 %	[119]
GHS/MWCNTs-CT	Natural cotton thread (CT)	hydrothermal process and dip coating	Polymer gel	PVA/H ₃ PO ₄	97.73 μ F/cm at 2 mV/s	3.06 $\times 10^{-3}$ μ Wh/cm	1.25 μ W/cm	8000	95.51 %	[120]
Reduced GO coated cotton fabric	Cotton fabric	Coating	Polymer gel	PVA/H ₃ PO ₄	464 F/g at 0.25 A/g	27.05 W h/kg	N.R.	1000	91.6 %	[79]
rGO-cotton electrode	Cotton woven	Screen printing of GO, followed by electrochemical reduction	Polymer gel	PVA-H ₂ SO ₄	2.5 mF/cm ² , 257 F/g	N.R.	N.R.	10,000	97 %	[121]
Reduced graphene oxide coated carbon fibers	Carbon fibers	Dip coating	Polymer gel	PVA/H ₃ PO ₄	13.5 mF/cm (307 mF/cm ²) at 0.05 mA/cm	1.9 μ Wh/cm, 21.4 μ Wh/cm ²	748.6 μ W/cm, 8.5 mW/cm ²	5000	85 %	[122]
Reduced graphene oxide on textile fabrics	Polypropylene fabric	Reactive inkjet printing (RIP) technique	Polymer gel	PVA/H ₃ PO ₄	13.3 m F/cm ² (79.9 F/g) at 0.1 mA/cm ²	1.18 mWh/cm ²	4.6 mW/cm ²	5000	~100 %	[123]
RGO/cotton fabric	Cotton cloth	brush-coating and drying process	Organic	2MEMIMBF ₄ /Acetonitrile	73.2 F/g at 0.1 A g	12.3 Wh/Kg	N.R.	1500	93.8 %	[124]

(continued on next page)

Table 2 (continued)

Electrode material	Flexible substrate	Fabrication process	Electrolyte type	Electrolyte material	Capacitance	Energy density	Power density	Cycling stability		Ref.
								Cycles	Capacitance retention (%)	
rGO–Ni-coated textile	Polyester/nylon textile	Hydrothermal reaction and dip coating	Aqueous	6 M KOH	81.7 F/g at 10 mV/s 8.19 mF/cm ² at 0.01 V/s	7.13 Wh/kg 0.28–0.51 μWh/cm ²	1.5 kW/kg 2.4 mW/cm ²	10,000	91 %	[125]
rGO/CCF	Carbonized cotton fabric (CCF)	Dip coating and annealing process	Aqueous	6 M KOH	87.5 mF/cm ² at 2 mV/s	11.77 μWh/cm ²	124.99 μW/cm ²	1000	89.82 %	[126]
rGO/SFF	Silver fiber fabric (SFF)	Electrophoretic deposition	Aqueous	3 M KOH	172 mF/cm ² at 4 mA/cm ²	N.R.	N.R.	5000	97 %	[127]
Electrode material	Flexible substrate	Fabrication process	Electrolyte type	Electrolyte material	Capacitance	Energy density	Power density	Cycling stability		Ref.
								Cycles	Capacitance retention (%)	
SWNTs/rGO electrode	Polyester (PET) substrate	Hydrothermal processes	Gel	PVA/H ₃ PO ₄	116.3 mF/cm ² , 45 F/cm ³ at 26.7 mA/cm ³	6.3 mWh/cm ³	1085 mW/cm ³	10,000	93 %	[128]
GO/PET	Polyethylene terephthalate (pet) fabric.	Coating	Polymer gel	PVA- H ₂ SO ₄ gel	756 μ F/cm ² at 20 mV/s	53 μWh/cm ³	1.4 W/cm ³	1000	98.3 %	[129]
Graphene/cotton composite	Cotton	Dipping and drying process followed by reduction method	Aqueous	1 M Na ₂ SO ₄	40 F/g at 0.85 A/g	N.R.	N.R.	1000	90 %	[130]
Graphene-coated textile electrodes	Cotton fabric	Microfluidization and Dip coating	Polymer gel	PVA/H ₂ SO ₄	2.7 m F/cm ² (electrode)	N.R.	N.R.	15,000	98 %	[131]
Graphite pen ink	Carbon fiber and Gold coated plastic fiber	Dip-coating method	Polymer gel	PVA-H ₂ SO ₄	11.9–19.5 mF/cm ²	1.76–2.70 μWh/cm ²	Up to 9.07 mW/cm ²	15,000	~100 %	[132]
Graphene ink screen printed on cotton textiles	Cotton fabric	ink screen printing	Polymer gel	PVA/H ₂ SO ₄	3.2 mF/cm ²	0.28 mWh/cm ²	3 mW/cm ²	10,000	95 %	[133]
Graphene and silk fibroin based carbon material	Silk fibroin	Carbonization	Aqueous	KOH	256 F/g at 0.5 A/g	14.4 Wh/kg	40,000 W/kg	10,000	96.34 %	[134]
Graphene@ cotton	Cotton	Dip coating	Aqueous	1 M Na ₂ SO ₄	232 mF/cm ² at 1 mA/cm ²	4.38 μWh/cm ²	5 mW/cm ²	5000	94.2 %	[135]
Nylon-graphene nonwoven composite	Nylon fiber	Dip-coating	Polymer gel	PVA-H ₂ SO ₄	10.37 mF/cm ² at 10 mV/s	1.44 μWh/cm ²	78.37 mW/cm ²	–	–	[136]
Hydrothermally activated graphene fiber fabrics	Graphene fiber fabrics	Hydrothermal activation process	Polymer gel	PVA/H ₂ SO ₄	1060 mF cm ⁻² at 1 mA/cm ² 244F/g at 0.1A/g	23.5 μWh/cm ²	26.3 mW/cm ²	300	92.2 %	[137]
Reduced graphene/ carbon nanotube core-sheath fiber	Sodium carboxymethyl cellulose (CMC)	Wet-spinning assembly strategy	Aqueous	1 M H ₂ SO ₄	269 mF/cm ²	5.91 μWh/cm ²	N.R.	2000	~100 %	[69]
			Solid gel electrolyte	PVA/H ₃ PO ₄	177 mF/cm ²	3.84 μWh/cm ²	0.02 mW/cm ²			
Electrode material	Flexible substrate	Fabrication process	Electrolyte type	Electrolyte material	Capacitance	Energy density	Power density	Cycling stability		Ref.
								Cycles	Capacitance retention (%)	
CNT and graphene modified composite ACFE textile electrode	Activated Carbon fiber felt	Freeze-drying process.	Aqueous	KOH	3350 mF/cm ² , device 2700 mF/cm ²	112 μWh/cm ²	490 μW/cm ²	1000	No decay at 1000 Cycles	[138]
Graphene on Stainless steel fabrics (SSF)	Stainless steel fabric	Hydrothermal method	Aqueous	H ₂ SO ₄	730.8 mF/cm ² at 2 mA/cm ² , 180.4 mF/	19.2 W h/cm ²	386.2 W/cm ²	7500	96.8 %	[139]

(continued on next page)

Table 2 (continued)

Electrode material	Flexible substrate	Fabrication process	Electrolyte type	Electrolyte material	Capacitance	Energy density	Power density	Cycling stability		Ref.
								Cycles	Capacitance retention (%)	
TRGO/carbon fabric	carbon fabric	Dip coating and thermal treatment	Polymer gel	H ₃ PO ₄ /PVA	cm ² at 1 mA/cm ² 70.4 F/g at 5 mV/s 120 F/g, 37 mF/cm at 2 mV/s	5.8 W h/kg N.R.	27.7 kW/kg N.R.	1000	93 %	[140]
Yarns welded with activated carbon and twisting with stainless steel yarn	Cellulose yarns	Welding process and Coating	Polymer gel	PVA-H ₃ PO ₄				3000	77 %	[141]
Carbon nanotube/stainless steel core-sheath yarn	Core-sheath yarn	Spinning and coating	Ionic-liquid gel	PVDF-HFP/EMIMBF ₄ /PC	263.31 F/cm ³ 329.13 F/g at 0.1 V/s	66.7 mWh/cm ³	8.89 W/cm ³	1000	91.9 %	[142]

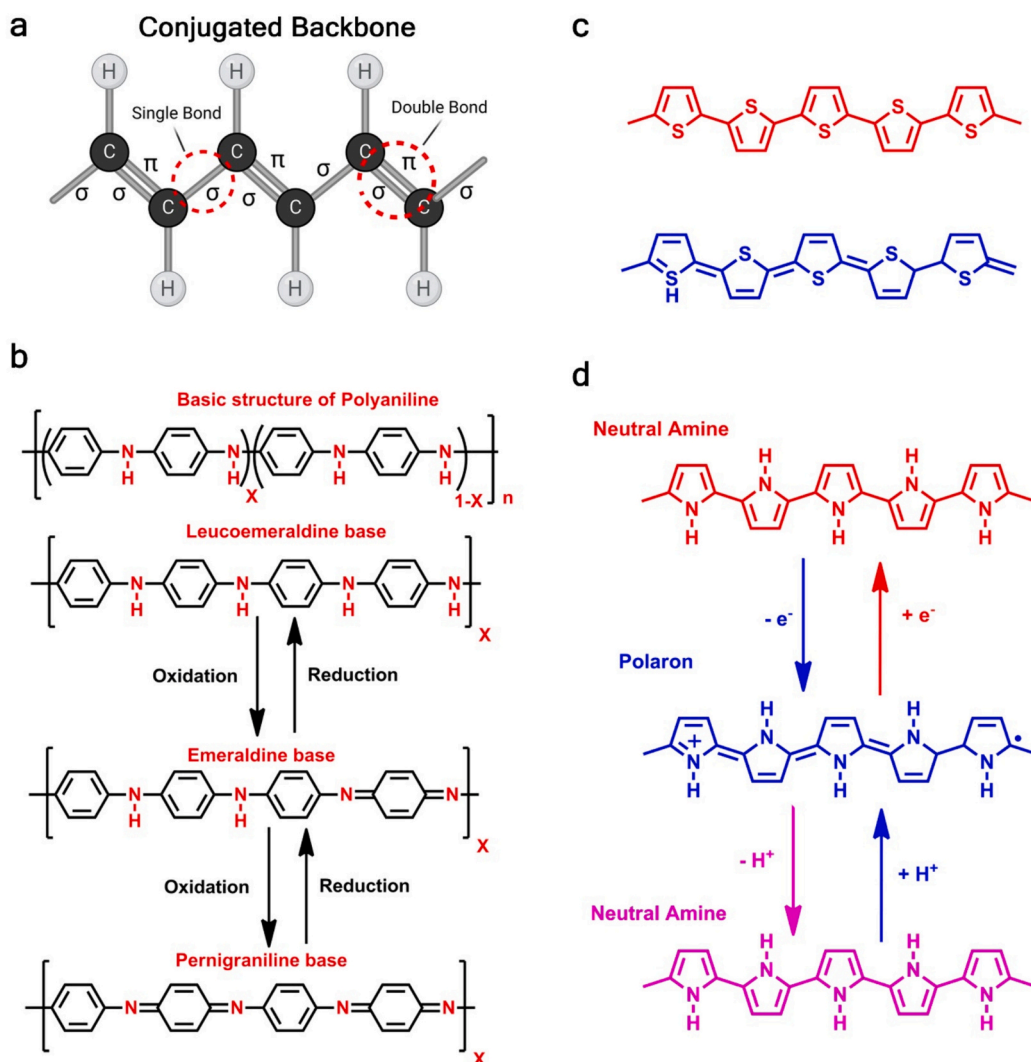


Fig. 3. a) Conjugated backbone of conductive polymers showing alternating pi and sigma single and double bonds; b) polyaniline basic, reduced, and oxidized forms, where X indicates the degree of reduction or oxidation, and n defines the polymer chain length; c) polythiophene basic and oxidized forms; d) polypyrrole neutral, oxidized, and reduced forms.

SSA, high electronic conductivity, excellent electrochemical properties, hydrophilicity, geometric topological structure, tunable terminations, excellent processability, and good thermal and mechanical stability [52,184]. MXenes are two-dimensional transition metal carbides, nitrides, and carbonitrides with a general chemical formula of $M_{n+1}X_nT$ where M signifies a transition metal, X is carbon or nitrogen, and T stands for the surface terminations. MXenes are derived from MAX

phases, where A represents elements from groups 13 and 14 of the periodic table by selectively removing the A layer from the MAX phases [185]. MXenes can reach a conductivity of up to 10,000 S/cm and have an internal surface area ranging from 20 to 100 m²/g depending on the synthesis and processing methods of MAX phases and MXenes [85,186,187]. The most studied MXene is titanium carbide (Ti₃C₂), having high conductivity, tunable surface terminations, acceptable

Table 3

Summary of the electrochemical performances of textile supercapacitors based on conductive polymers to date.

Electrode material	Flexible substrate	Fabrication process	Electrolyte type	Electrolyte material	Capacitance	Energy density	Power density	Cycling stability		Ref.
								Cycles	Capacitance retention (%)	
PANI @ Cotton	Carbon cloth (CC)	Self-stabilized dispersion polymerization (SSDP)	Organic redox-active electrolyte	H ₂ SO ₄ -hydroquinone mixture	437.8 F/g at 0.5 A/g 875.6 mF/cm ² at 1 mA/cm ²	21.89 Wh/kg 43.78 μWh/cm ²	N.R.	8500	98.3 %	[158]
PANI@TS-silk fabric	Silk fabric	Ternary solvent treatment and in situ polymerization	Aqueous	1 M H ₂ SO ₄	4091.43 mF/cm ² at 1 mA/cm ²	N.R.	N.R.	3000	99.54 %	[159]
CF/PANI// Functionalized CF	Carbon fiber thread	Electrochemical deposition and dip coating	Polymer gel	PVA/H ₃ PO ₄	5 F/cm ³ at 2 mA/cm ³ ; 4.5 F/cm ³ at 5 mA/cm ³	2 mWh/cm ³	10.22 W/cm ³	10,000	81 %	[160]
PPy coated cotton	Cotton fabric	In situ polymerization and coating	Aqueous	1 M H ₂ SO ₄	Knitted 4117, woven 2191, and nonwoven fabrics 2905 mF/cm ²	5.94 Wh/kg	259.55 W/kg	Stable within 5000 cycles	Stable	[161]
PPy-coated cotton fabrics	Cotton fabrics	In situ polymerization	Aqueous	2.0 M NaCl	325 F/g at 0.6 mA/cm ²	24.7 Wh/kg	140.5 W/kg	500	63 %	[162]
PPy-coated cotton fabric	Cotton	In situ chemical polymerization	Aqueous	2.0 M NaCl	225 F/g at 0.6 mA/cm ²	N.R.	N.R.	200	92 %	[163]
PPy nanotubes on the cotton yarns	Cotton yarns	In situ polymerization	Polymer gel	PVA/H ₂ SO ₄	74.0 mF/cm ² at 0.42 mA/cm ²	7.5 μWh/cm ²	22.4 μW/cm ²	N.R.	N.R.	[164]
PPy-coated fabric	Cotton fabric	In situ polymerization	Polymer gel	LiCl/PVA Electrode	5073 mF/cm ² at 1 mA/cm ²	102.4 μWh/cm ²	0.39 mW/cm ²	2000	90 %	[165]
				LiCl/PVA device	1167.9 mF/cm ² at 1 mA/cm ²					
PPy layers on textile	Polypropylene non-woven textile	Reactive inkjet printing	Polymer gel	PVA/H ₂ SO ₄	72.3 F/g at 0.6 A/g	6.12 Wh/kg	139 W/kg	55.4 %	2000	[166]
CT/PPy	Carbon thread (CT)	Polymerization method.	Simulated sweat solution	Simulated sweat solution	2.3 F/g at 1.1 V	386.5 mWh/kg	46.4 kW/kg	1000	100 %	[167]

Electrode material	Flexible substrate	Fabrication process	Electrolyte type	Electrolyte material	Capacitance	Energy density	Power density	Cycling stability		Ref.
								Cycles	Capacitance retention (%)	
Polypyrrole/carbonized cotton fabric (PPy/CCF)	Carbonized cotton fabric	Carbonization and in-situ electrodeposition method	Polymer gel	LiCl/PVA	3596 mF cm ⁻² at 2 mA cm ⁻²	1.18 mWh/cm ³	17 mW/cm ³	4000	96.5 %	[168]
PEDOT: PSS-coated PET fabric	PET fiber	Dyeing and spray-coating method	Aqueous	1 M H ₂ SO ₄	46.18 m F/cm ² at 1 mA/cm ²	4.10 mW h/cm ²	400.03 mW/cm ²	200	87 %	[169]
PEDOT coated poly ester fiber	polyester fabric	In situ Polymerization	Aqueous	1 M Na ₂ SO ₄	0.64 F/cm ² at 2 mA/cm ² (80.8 F/g)	0.23 mWh/cm ³	N.R.	50,000	100 %	[170]
PEDOT-Cl/stainless steel yarns Sewn	Stainless steel threads	Coating and reactive vapor deposition	Polymer gel	PVA/H ₂ SO ₄	80 mF/cm ² (2 mV/s)	11 μW h/cm ²	N.R.	4000	71 %	[171]
Multifunctional polydopamine	Cellulosic textile	In situ polymerization	Aqueous	1 M H ₂ SO ₄	1208.4 mF/cm ² at 1 mA/cm ²	N.R.	N.R.	4000	94 %	[172]

oxidative stability among MXenes, fast surface redox reactions, and lack of toxicity when worn on the skin [7,188,189]. MXenes tend to oxidize into metal oxides quickly, so any flexible supercapacitors fabricated using these materials have a short lifespan and will produce more waste. This fact limits their applicability in wearable technology and flexible supercapacitors, and as a result, they are considered for short-term military and medical applications [190]. Thus, the recyclability of MXene-based wearable TSCs has been considered a challenge. Along these lines, a popular substrate for MXenes is a cellulose-based fiber that

can be dissolved and wet-spun into fibers. Cotton fiber coated with oxidized MXenes could be dissolved into spinning dope impregnated with the generated metal oxides and wet-spun into cellulose fibers to be recycled into wearable supercapacitors. This would also streamline the process by eliminating the need to synthesize conductive materials for this purpose and eliminate the need to coat existing yarn with the conductive materials [7,191]. The electrochemical performances of textile supercapacitors based on metal oxides are summarized in Table 4.

Table 4

The electrochemical performances of textile supercapacitors prepared with metal oxides, carbides, nitrides, and sulfides.

Electrode material	Flexible substrate	Fabrication process	Electrolyte type	Electrolyte material	Capacitance	Energy density	Power density	Cycling stability		Ref.
								Cycles	Capacitance retention (%)	
MnO ₂ /Carbonized cotton textile	Carbonized cotton textile	Carbonization and hydrothermal method	Aqueous	1 M Na ₂ SO ₄	465 F/g at 0.1 A/g	N.R.	N.R.	5000	95 %	[192]
MnO ₂ /carbon fabric	Carbonized silk fabric	Carbonization and hydrothermal	Aqueous	6 M KOH	105 f/g at 0.25 A/g	14.58 Wh/Kg	0.025 kW/kg	10,000	101.54 %	[193]
MnO ₂ -coated hollow carbon microspheres	Silk fabric	Screen-printing techniques	Polymer gel	PVA/H ₃ PO ₄	19.23 mF/cm ² at 1 mA cm ⁻²	N.R.	N.R.	2000	84 %	[80]
MnO ₂ /ACT hybrid composite.	Activated carbon textiles from cotton t-shirt	Electrochemical deposition	Aqueous	1 M Na ₂ SO ₄	120 F/g at 1 mA/cm ²	66.7 Wh/kg	4.97 kW/kg	1000	97.5 %	[194]
MnO ₂ nanoribbons and cotton-derived carbon cloth	Cotton-derived carbon cloth (CDCC)	Hydrothermal method	Aqueous	1 M Na ₂ SO ₄	202 mF/cm ² at 0.1 mA/cm ²	30.1 μWh/cm ²	0.15 mW/cm ²	5000	87.7 %	[195]
MnO ₂ @carbonized cotton textile (MCCT)	Cotton textile	Hydrothermal and carbonization method	Aqueous	1 M Na ₂ SO ₄	526.25 mF/cm ² (751.78 F/g) at 1 mA/cm ²	0.057 mWh/cm ² (5.71 mWh/cm ³)	39.71 mW/cm ³ (3.97 W/cm ³)	10,000	99.7 %	[196]
MnO ₂ /Carbon NPs/ carbon fabric	Carbon fabric	Simple flame synthesis method and electrochemical deposition process	Polymer gel	PVA/H ₃ PO ₄	109 mF/cm ² (165 F/g at 5 mV/s)	4.8 Wh/kg	14 kW/kg	10,000	97.3 %	[197]
H-TiO ₂ @MnO ₂ // H-TiO ₂ @C core-shell nanowires	Carbon fiber	Hydrothermal and deposition	Polymer gel	PVA/LiCl	141.8 F/g, 710 mF/cm ³ at 10 mV/s	0.3 mWh/cm ³ (59 Wh/kg)	230 mW/cm ³ (45 kW/kg)	5000	91.2 %	[198]
MnO ₂ @TiN on Carbon Cloth	Carbon cloth	Hydrothermal method and coating	Aqueous	5 M LiCl	2.69 F/cm ³ 756 mF/cm ² at 6 mA/cm ²	1.5 mWh/cm ³	1.71 W/cm ³	70,000	No decay at 70000 cycles	[199]
NiO/MnO ₂ /CC	Carbon cloth (CC)	Hydrothermal and annealing treatment	Aqueous	1 M Na ₂ SO ₄	316.37 mF/cm ² and 204.3 F/g at 50 mV/s	N.R.	N.R.	2200	89 %	[200]
Nickel di-selenide (NiSe ₂)/carbon cloth	Carbon cloth (CC)	Hydrothermal technique	Polymer gel	PVA/KOH	980 F/g at 1A/g	50 Wh/kg	900 W/kg	25,000	98 %	[201]
NiWO ₄ /NiO _x /CCF	Commercial cotton fabrics	Spray coating method	Polymer gel	PVA/KOH	135 mF/cm ² at 0.2 mA/cm ²	12 μWh/cm ²	69 μW/cm ²	5500	80 %	[202]
Cotton/Ni/Co-Ni LDH hybrid yarn	Cotton thread (CT)	Solvothermal method	Polymer gel	KOH/PVA	221 mF/cm ² at 0.04 mA/cm ²	9.3 μWh/cm ²	43.99 μW/cm ²	1200 2000	82.5 % 79 %	[203]
Electrode material	Flexible substrate	Fabrication process	Electrolyte type	Electrolyte material	Capacitance	Energy density	Power density	Cycling stability		Ref.
								Cycles	Capacitance retention (%)	
(FeCo ₂ S ₄ -NiCo ₂ O ₄) ternary metal sulfides composites	Silver-sputtered textile cloth	Coating, silver-sputtering, and sulphuration	Polymer gel	PVA/KOH	1519 F/g at 5 mA/cm ²	46 W h/kg	1070 W/kg	3000	92 %	[204]
NiCo ₂ S ₄ nano-arrays	Activated carbon textile	Hydrothermal method	Aqueous	3 M KOH	183.2 mA h/g at 2 mA/cm ²	56.2 Wh/kg	632.7 W/kg	1000	82.6 %	[205]
(NiCo ₂ S ₄ @NiCo ₂ O ₄) nanocolumn arrays	Carbon textile	Hydrothermal method and electrodeposition	Aqueous	3 M KOH	2258.9 F/g at 0.5 A/g	44.06 Wh/kg	6.4 kW/kg	6000	92.5 %	[206]
Ternary nickel cobalt sulfide compound (Ni-Co-S nanoflakes)	Cotton textile	Electrochemical deposition	Solid state electrolyte	PVA/KOH	1098 F/g at 1 mA/cm ²	48.9 Wh/kg	390 W/kg	5000	70 %	[207]
NiCo ₂ O ₄ @NiCo ₂ O ₄ /ACT	Cotton activated carbon textile (ACT)	Hydrothermal, calcination process and coating	Polymer gel	PVA/KOH	1929 F/g at 1 mA/cm ²	83.6 Wh/kg	8.4 kW/kg	N.R.	N.R.	[208]
NiFe ₂ O ₄ (NFO) on carbon textile	carbon textile	Hydrothermal Method	Polymer gel	LiCl-PVA	584 F/g at 5 mV/s	54.9 W h/kg	300 W/kg	10,000	93.57 %	[209]

(continued on next page)

Table 4 (continued)

Electrode material	Flexible substrate	Fabrication process	Electrolyte type	Electrolyte material	Capacitance	Energy density	Power density	Cycling stability		Ref.
								Cycles	Capacitance retention (%)	
Ni/Cu-coated polyester fiber	polyester yarns	Hydrothermal and dip-coating	Polymer gel	PVA/KOH	385.4 mF/cm ² at 1 mA/cm ²	78.1 μ Wh/cm ²	14 mW/cm ²	5000	82.7 %	[210]
ZnO @ amorphous ZnO-doped MnO ₂ on carbon cloth	Carbon cloth	Wet chemical process and Dip coating	Polymer gel	PVA/LiCl	26 mF/cm ² (325 mF/cm ³) at 0.5 mA/cm ²	0.04 mWh/cm ³	2.44 mW/cm ³	10,000	87.5 %	[211]
ZnO@CFT	Carbon fiber textile (CFT)	Hydrothermal process	Aqueous	1 M KOH	201 F/g at 1 A/g	N.R.	N.R.	3000	90.32 %	[212]
Zinc sulfide (ZnS)/Carbon textile (CT)	Carbon textile	Hydrothermal process	Polymer gel	LiCl – PVA	540 F/g (56.25 F/cm ²) at 5 mV/s	51 W h/kg	205 W/kg	5000	94.6 %	[213]
(Mn ₃ O ₄ @NPC)//PC	Carbon cloth	Annealing and hydrothermal processes	Polymer hydrogel	PVA/Na ₂ SO ₄	81.97 F/g at 1 A/g	32.65 Wh/kg	26.02 kW/kg	10,000	92.71 %	[214]
(Mn ₃ O ₄) nanowalls/carbon fiber	carbon fiber (CF)	electrochemical deposition	aqueous	1 M Na ₂ SO ₄	300.7 F/g 571 mF/cm ² at 5 mV/s	41.7 Wh/kg	N.R.	7500	100 %	[215]
NiMnO ₃ nanosheets on carbon cloth	Carbon cloth (CC)	hydrothermal method	aqueous	6 M KOH	2330 F/g at 1 A/g	N.R.	N.R.	1000	67.8 %	[216]
Electrode material	Flexible substrate	Fabrication process	Electrolyte type	Electrolyte material	Capacitance	Energy density	Power density	Cycling stability		Ref.
								Cycles	Capacitance retention (%)	
MnO ₂ on carbon nanoparticle coated carbon fiber	Carbon fiber	Electrodeposition and dip coating	Solid-state electrolyte	LiCl-PVA	4.6 F/cm ³ at 0.25 mA/cm ³	2.1 Wh/cm ³	8 W/cm ³	10,000	81.2 %	[217]
(N-C) nanowire/(Fe ₂ O ₃ and MnO ₂) nanocomposite into carbon fabric.	Carbon fabric	Electropolymerization and post carbonization	Gel electrolyte	LiCl/PVA	72 mF/cm ² 0.5 mA/cm ²	N.R.	N.R.	5000	92 %	[218]
Co-MoS ₂ nanoflower	Carbon fabric	Hydrothermal method	Aqueous	1 MKOH	86 F/g	4.30 Wh/kg	0.6 KW/kg	10,000	98.5 %	[219]
Co ₃ O ₄ nanowires on carbon fabric	Carbon fabric	Hydrothermal method	Polymer gel electrolyte	PVA-KOH	3290 F/g at 5 mV/s	6.7 Wh/kg	5 kW/kg	5000	95.3 %	[220]
(MoS ₂) nanosheets on carbon fabrics	Carbon cloth	Hydrothermal method	Aqueous	1 M LiOH	159.38 mF/cm ² at 0.5 mA/cm ²	N.R.	N.R.	15,000	80.6 %	[221]
Pt-doped MoS ₂ /carbon cloth	Carbon cloth	Hydrothermal method	Polymer gel	PVA/H ₃ PO ₄ electrode PVA/H ₃ PO ₄ (device)	250 F/g at 0.5 A/g 42 F/g at 0.4 A/g	N.R.	N.R.	3000	87.96 %	[222]
MXene/carbonized silk fabric	Silk-derived carbon fiber cloth	Carbonization and deposition	Aqueous	1 M H ₂ SO ₄	362 mF/cm ² at 2 mV/s	13 μ Wh/cm ²	181 μ W/cm ²	1000	96 %	[223]
3D knitted cotton yarn coated/MXene	Cotton yarn	Dip-coating process	Polymer gel	H ₃ PO ₄ /PVA	519 mF/cm ² at 2 mV/s	25.4 μ Wh/cm ²	0.47 mW/cm ²	10,000	~100 %	[7]
MXene-coated cotton fabric (MCF)	Cotton fabric	Dip coating process	Aqueous	1 M H ₂ SO ₄	208 mF/cm ² at 5 mV/s	2.01 μ Wh/cm ²	5.3 mW/cm ²	10,000	~ 96.1 %	[224]
MXene nanofiber @PET yarn	Polyester (PET) yarn	Electrospinning	Polymer gel	PVA/H ₂ SO ₄	18.39 mF/cm ² at 5 mV/s	0.38 μ Wh/cm ²	0.39 mW/cm ²	6000	98.2 %	[225]
Carbonized MXene/Cotton fabric	Carbonized cotton fabric	Carbonization process and dip-coating	Polymer gel	PVA-H ₃ PO ₄	794.2 mF/cm ² (233.6 F/g) at 2 mV/s	N.R.	N.R.	10,000	74 %	[226]
MXene coated woven cotton textile	woven cotton textile	Dip coating and drop casting method	Polymer gel electrolyte	LiCl/PVA	146 mF cm ⁻² at 0.16 mA cm ⁻²	0.401 mW h cm ⁻²	0.401 mW h cm ⁻²	N.R.	N.R.	[227]
MXene@ textile	woven fabric	screen-printing strategy	Polymer gel electrolyte	H ₂ SO ₄ /PVA	4979.2 mF cm ⁻²	62.2 μ Wh cm ⁻²	N.R.	3365	80.53 %	[228]

3.4. Hybrid materials

Hybridization of active materials from one or more subgroups is one of the most important methods for fabricating high-performance textile-based supercapacitors. These components can be used together to create composite electrodes [52,99]. Many researchers worldwide have focused on hybridizing carbon components with conductive polymers or metal oxides to manufacture high-performance textile-based supercapacitors. Table 5 reviews the electrochemical performances of textile supercapacitors based on hybrid materials.

For instance, carbonaceous and metallic substrates can be coated with conductive polymers. Applying thin films of polymer on these supports suppresses charge trapping and irreversibility induced by swelling and shrinking during charge and discharge. The conductive polymer components in these composites provide a few other advantages: their inherent chemical stability and flexibility, which dovetail with the required flexibility of TSCs [229]. The carbonaceous or metallic materials provide a higher SSA and cycling longevity because of their porosity and EDLCs behavior. Researchers have coated these materials with conductive polymers to introduce pseudocapacitive behavior into TSCs made with substances that store energy primarily via EDLCs as the main electrode material. The carbonaceous/metallic primary electrode material supports the conductive polymer framework while it swells and shrinks during charge and discharge. Meng et al. coated PANI onto CNT to create flexible paper electrodes separated by a sulfuric acid/PVA electrolyte and achieved a 1.3 mF/mm^2 specific capacitance with 94 % retention after 10,000 cycles [230]. In contrast, Table 2 shows that CNTs by themselves or with other carbonaceous materials generally have capacitance retention closer to 90 % with lower longevity.

Transition metal carbides such as MXenes can have conductive polymers polymerized directly on their surface because their oxide surface terminations act as a catalyst for the polymerization of conductive polymers. Boota et al. [229] created MXene/Ppy flexible electrodes through oxidant-free polymerization of Ppy on MXene with 1000 F/cm^3 specific capacitance and 92 % retention after 25,000 cycles. VahidMohammadi et al. [231] created freestanding MXene/PANI electrodes that achieved a gravimetric capacitance of 503 F/g with 98 % retention after 10,000 cycles.

4. Electrolyte materials for wearable textile-based supercapacitors

Like electrodes, electrolytes are essential in wearable supercapacitors (SCs). They are important from both safety and performance perspectives. Both solid and liquid electrolytes are used in flexible and wearable supercapacitors. Electrolyte materials are critical in shifting and balancing charges between the two electrodes [346]. The choice of electrolyte affects the electrochemical performance of textile supercapacitors by influencing ion conductivity, the electrochemical stable potential window (ESPW), equivalent series resistance (ESR), chemical and thermal stabilities, operating temperature range, and compatibility with electrode materials [346,347]. The type, composition, and concentration of the electrolyte are important parameters for fabricating high-performance supercapacitors [52].

Widening the electrolyte potential window can effectively increase the energy density and performance of TSCs because both energy and power densities are proportional to the square of the cell voltage [346]. Therefore, in TSC research, developing new electrolyte solutions with wide potential windows should be given even higher priority than developing new electrode materials [86,346]. Besides the intrinsic properties of the electrolyte potential window, the compatibility or the possible interaction between the electrolyte, electrode materials, and inactive components, including current collectors, binders, and separators, is very important in achieving high-performance SCs. Depending on the type and nature of the electrolyte, inactive components and electrode materials may profoundly influence the performance and

reliability of the TSCs [346,348]. Ion concentration and mobility play important roles in electrolyte ionic conductivity. Electrolytes with high ionic conductivities are essential for achieving high power density and fast charge/discharge cycles. The value of the ESR also affects the performance of TSCs because it is directly related to the electrolyte's ionic conductivity and can have a strong effect on the power density [349]. High ESR (or low ion conductivity) limits the charging/discharging rate, resulting in lower power and energy density.

The cycle life of the TSCs depends on many factors, such as the cell type, electrode material, electrolyte, charging/discharging rate, operating voltage, and temperature [346,350]. Additionally, the presence of impurities in the electrolytes affects the performance of TSCs and influences the ESPW and self-discharge rate of TSCs [350,351]. The aging and failure of TSCs are related to the decomposition of the electrolytes caused by cell voltage, the electrolyte ion intercalation, and harsh working conditions such as extreme temperatures or stress/strain of the TSCs [346,352,353]. These electrolyte properties, in turn, impact the equivalent circuit parameters, such as ESR, double-layer capacitance, Faradaic resistance, and Warburg impedance, thereby determining the overall efficiency, power density, and energy density of the supercapacitor. Selecting an appropriate electrolyte that minimizes the ESR is thus critical for optimizing the performance of textile supercapacitors.

To date, an ideal electrolyte that meets all the requirements of an ideal electrolyte has not been developed. For developing an ideal electrolyte, numerous criteria have been developed: wide potential window, high ionic conductivity, high chemical and electrochemical stability, inert to other TSC components (e.g., electrodes, current collectors, binders, and separators), wide operating temperature range, low volatility and flammability, inexpensive, and environmentally friendly [346,347]. Each electrolyte has its advantages and disadvantages. For example, supercapacitors containing solid electrolytes are less prone to leakage problems associated with liquid electrolytes, but they suffer from low conductivity. Aqueous electrolytes have high conductivity and ionic capacity but have limited working voltage, leading to a lower energy density, lower cycle stability, and leakage problems. Organic and ionic liquid electrolytes can operate at higher voltages but have lower ionic conductivity. The need for an electrolyte with a high ionic conductivity, cycle stability, and wide potential window has stimulated tremendous research efforts to improve the overall performance of electrolytes in TSCs and to pursue new formulations for electrolytes. The common electrolytes are mainly classified as liquid and solid/quasi-solid-state electrolytes [348]. The different types of electrolytes used in supercapacitors are given in Tables 2, 3, 4, and 5. Here we will discuss each of the various types of electrolytes in more detail.

4.1. Liquid electrolytes

Liquid electrolytes perform better in energy storage than solid or semi-solid/gel electrolytes because they are liquid-based electrolytes with low viscosity and high ionic conductance. However, integrating liquid electrolytes into wearable textiles presents leakage, stability, and comfort challenges. Liquid electrolytes are classified into aqueous electrolytes and non-aqueous electrolytes (organic electrolytes and ionic liquids (ILs) electrolytes) [346,347].

Aqueous electrolytes have been the subject of extensive research. This is mainly because aqueous electrolytes are inexpensive and can be easily prepared in the laboratory without strict preparation procedures. The maximum working voltage of aqueous electrolyte is limited to 1.23 V owing to the thermodynamic decomposition of water [351]. Aqueous electrolytes can be categorized into acid (such as H_2SO_4 solution-representative of strong acid aqueous electrolytes and the most frequently used in this category), alkaline (such as KOH solution-representative of strong base aqueous electrolytes, and the most frequently used in this category), and neutral (such as Na_2SO_4 solution-representative of all neutral aqueous electrolytes and the most frequently used in this category) solutions. Aqueous electrolytes have

Table 5

The electrochemical performances of textile supercapacitors based on hybrid materials to date.

Electrode material	Flexible substrate	Fabrication process	Electrolyte type	Electrolyte material	Capacitance	Energy density	Power density	Cycling stability		Ref.
								Cycles	Capacitance retention (%)	
Textile supercapacitors based on MXene and other materials										
MXene/PANI@ Cotton	Cotton	In-situ polymerization and dip-coating	Polymer gel electrolyte	PVA-H ₂ SO ₄	1027.5 mF/cm ² at 1 mA/cm ²	N.R.	N.R.	10,000	61.7 %	[232]
PANI/MXene@ PET fabric	PET fabric	In-situ polymerization and spray-coating	Polymer gel electrolyte	H ₂ SO ₄ /PVA	647 mF/cm ² at 0.3 mA/cm ²	8.08μWh/cm ² 0.69 Wh/kg	0.045 mW/cm ² 3.8 W/kg	–	–	[233]
PANI/MXene (V ₂ C)	Cotton	In-situ polymerization	Polymer gel electrolyte	H ₂ SO ₄ /PVA	337.5 F/g at 1 A/g	11.25 Wh/kg	415.38 W/kg.	10,000	97.6 %	[234]
MXene/PEDOT-PSS coated CF	Carbon fibers	Dip coating	Polymer gel electrolyte	PVA/H ₃ PO ₄	658.5 mF/cm ² 131.7 mF/cm at 0.2 mA/cm	N.R.	N.R.	10,000	~90 %	[41]
PPy/MXene@Cotton fiber	Cotton fiber	In-situ polymerization	Aqueous	1 M H ₂ SO ₄	455.9 mF/cm ² at 0.9 mA/cm ²	N.R.	N.R.	2000	83.3 %	[235]
Polypyrrole – MXene coated textile	Cotton textile	dipping-coating method	Polymer gel electrolyte	H ₂ SO ₄ /PVA	343.20 F/g at 2 mA/cm ²	1.30 mW h/g	41.1 mW/g	2000	~80 %	[236]
PEDOT/MXene decorated cotton fabrics	Cotton fabrics	Vapor phase polymerization and spray-coating strategy	Polymer gel electrolyte	H ₂ SO ₄ /PVA	1000.2 mF/cm ² at 0.5 mA cm ²	12.5 μWh/cm ²	N.R.	N.R.	N.R.	[237]
MXene/PEDOT: PSS hybrid fiber	Fiber	wet-spinning approach	Polymer gel electrolyte	PVA/H ₂ SO ₄	614.5 F/cm ³ at 5 mV/s	≈7.13 Wh/cm ³	≈8249 mW/cm ³	10,000	95 %	[238]
rGO/MXene hybrid fiber	Graphene fiber	Hydrothermal strategy	Aqueous	1 M H ₂ SO ₄	345 F/cm ³ 195 F/g at 0.1 A/g	30.7 mWh/cm ³	70.7 mW/cm ³	7500	124.8 %	[239]
CNTs/MXene-TPU hybrid fiber	Thermoplastic polyurethane (TPU) fiber	Wet spinning technique	Polymer gel electrolyte	PVA/H ₂ SO ₄ gel	3.9 F/cm ³ at 1 V/s	1.16 mWh/cm ³	0.16 W/cm ³	N.R.	N.R.	[240]
MXene-decorated NCFT (MNCFT) and ONCFT electrodes (ONCFT// MNCFT)	N-doped carbon fiber textile (NCFT) substrate	Chemical oxidization and MXene ink painting	Polymer gel electrolyte	PVA/H ₂ SO ₄	780 mF/cm ² at 1 mV/s	277.3 μWh/cm ² (16.3 Wh/kg)	36.7 W/kg	30,000	90 %	[241]
MXene/silver@ nylon yarn	Silver-plated nylon fibers	Coating	Polymer gel electrolyte	PVA/H ₂ SO ₄	328 mF/cm ² at 2 mV/s	7.3 μWh/cm ²	132 μW/cm ²	10,000	~100 %	[242]
MXene/RuO ₂ coated carbon fabric	Carbon fabric (CF)	Hydrothermal and Coating	Aqueous	1 M H ₂ SO ₄	416 mF/cm ² , 200 F/g	37 μWh/cm ²	40 mW/cm ²	20,000	86 %	[243]
Electrode material	Flexible substrate	Fabrication process	Electrolyte type	Electrolyte material	Capacitance	Energy density	Power density	Cycling stability		Ref.
								Cycles	Capacitance retention (%)	
Textile supercapacitors based on MXene and other materials										
NiCo ₂ S ₄ @W-MX/CF	Carbon fiber (CF)	Wet-spinning method and electrodeposition	Polymer gel electrolyte	PVA/KOH	2160 mF/cm ² at 3.86 mA/cm ²	40.70 mWh/cm ³	301.51 mW/cm ³	10,000	85.07 %	[244]
MXene/cellulose nanofiber anode and carbon cloth/ polyaniline cathode Ti ₃ C ₂ /CNF// PANI/CC	Carbon cloth	Vacuum filtration and in situ polymerization method.	Polymer gel electrolyte	PVA/ H ₂ SO ₄	452 mF/cm ² at 5 mV/s	30.6 Wh/kg	1211 W/kg	5000	86 %	[245]
Carbon cloth/ MXene PANI/ CoNi-LDH composite	Carbon cloth	Electrodeposition and coating	Polymer gel electrolyte	PVA-KOH	1200 F/g at 1 A/g	39.33 Wh/kg	399.95 W/Kg	10,000	91 %	[246]

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Table 5 (continued)

Electrode material	Flexible substrate	Fabrication process	Electrolyte type	Electrolyte material	Capacitance	Energy density	Power density	Cycling stability		Ref.
								Cycles	Capacitance retention (%)	
MXene@ PANI coated activated carbon cloth	Activated carbon cloth (ACC)	Polymerization and dip-coating	Aqueous	1 M H ₂ SO ₄	1347 mF/cm ² at 1 mA/cm ²	370 μWh/cm ²	2.33 mW/cm ²	5000	81 %	[247]
MXene/AgNWs-decorated textiles	Nonwoven fabrics	Spray coating method	Polymer gel electrolyte	PVA-KOH	2390mF/cm ² at 1.2 mA cm ⁻²	119.5 μWh/cm ²	0.36 mW/cm ²	200	~100 %	[248]
RGO/ MXene@cotton fabric	Cotton fabric	Dip-coating and spray-coating method.	Polymer gel electrolyte	H ₂ SO ₄ / PVA	383.3 F/g (258 mF/cm ²) at 5 mV/s	N.R.	N.R.	N.R.	N.R.	[249]
RGO@ MXene yarn	fiber	Dip-coating	Polymer gel electrolyte	H ₂ SO ₄ / PVA	253.01 mF/cm ² at 20 mV/s	27.1 μWh/cm ²	2502.6 μW/cm ²	1000	90 %	[250]
MXene@RGO/ MXene@PEDOT: PSS	fiber	Wet-spinning method	Polymer gel electrolyte	H ₂ SO ₄ / PVA	53.1 F/cm ³ at 50 mA/cm ³	16.6 mWh/cm ³	37.5 mW/cm ³	5000	96.6 %	[251]
MWCNT- MXene@carbon cloth	Carbon cloth	CVD method and electrodeposition	Polymer gel electrolyte	PVA- H ₂ SO ₄	114.58 mF/cm ² at 1 mA/cm ²	22.11 mWh/cm ³	2.99 W/cm ³	16,000	92 %	[252]
MXene/GO fibers	fiber	Wet-spinning process	aqueous	H ₂ SO ₄	890.7 F/cm ³ , 494.8 F/g at 10 mV/S	N.R.	N.R.	1000	80 %	[253]
PANI/ carbon@textile	Cotton textile	Polymerization, carbonization	Polymer gel electrolyte	PVA/ H ₂ SO ₄	386.7 mF/cm ² at 1 mA/cm ²	35.8 mWh/ m ²	745 mW/ m ²	5000	70 %	[254]
ACFF/PANI/CNT composite	Activated carbon fiber felt (ACFF)	Polymerization and coating	aqueous	1 M H ₂ SO ₄	5611 mF/cm ² at 2 mA/cm ²	185 μWh/ cm ²	4517 μW/ cm ²	5000	85 %	[255]

Electrode material	Flexible substrate	Fabrication process	Electrolyte type	Electrolyte material	Capacitance	Energy density	Power density	Cycling stability		Ref.
								Cycles	Capacitance retention (%)	
Textile supercapacitors based on MXene and other materials										
PANI/CNT/CNF	Carbon nanofibers (CNF)	Electrospinning, CNT growth, and polymerization.	Aqueous	1 M H ₂ SO ₄	503 F/g at 0.3 A/g	70 Wh/ kg	15 kW/ kg	1000	92 %	[256]
CNT/PANI composite	CNT fiber textile	Chemical vapor deposition	Polymer gel electrolyte	PVA/H ₃ PO ₄	272.7 F/g at 1A/g	N.R.	N.R.	2000	90 %	[257]
PANI/N-CNT@CNT fiber	CNT fiber	CVD process and electrochemical deposition	Aqueous	1 M H ₂ SO ₄	323.8 F/g at 1 A/g	5.9 W h/kg	0.2 kW/kg	10,000	92.1 %	[258]
PANI/CNT/EVA composite @ cotton	Cotton material	Electrodeposition method	Polymer gel electrolyte	PVA/H ₂ SO ₄	620.1 mF/cm ² at 0.1 mA/cm ²	26.7 mWh/ cm ²	100 mW/ cm ²	3000	66.4 %	[259]
PANI/CNT@ Pineapple-polyester woven blended fabric	PPWF fabric	Dip-and-dry process and in situ polymerization	Aqueous	1 M H ₂ SO ₄	386 mF/cm ² at 1 mA/cm ²	N.R.	N.R.	1600	55.2 %	[260]
Elastic fiber/CNTs/ PANI	Elastic fiber substrate	Electro-deposition	Polymer gel electrolyte	PVA/H ₃ PO ₄	255.5 F/g at 1 A/g	12.75 W h/kg	1494 W/kg	10,000	69 %	[261]
CNT@PANI yarn	CNT yarn	CVD method and in-situ polymerization	Polymer gel electrolyte	PVA/H ₂ SO ₄	38 mF/cm ² at 0.01 mA/cm ²	N.R.	N.R.	800	91 %	[56]
MWCNTs/PANI/ cotton fabric	Cotton	Dip coating	aqueous	2 M Na ₂ SO ₄	590.93 F/g at 0.001 V/s	N.R.	N.R.	3000	89 %	[262]
PANI@MWCNT	Nickel foam	CVD and in situ polymerization	aqueous	1 M KOH	296 F/g at 1.6 A/g	1 J/m ²	0.3 W/ m ²	2000	95 %	[263]
MWCNT–PANI composite	MWCNT fiber	Electrodeposition, polymerization	Polymer gel electrolyte	PVA/H ₃ PO ₄	274 F/g at 2 A/g	N.R.	N.R.	1000	99 %	[264]
PET/MWCNTs/ PANI	Polyester fabric	CVD method and polymerization	Polymer gel electrolyte	PVA/H ₃ PO ₄ strip-shaped electrode	421.7 F/cm ³ (343.6 F/g at 0.5 A/cm ³)	9.56 mWh/ cm ³	2.91 W/cm ³	10,000	Stable within 10,000 cycles	[265]
PANI/SWCNT/cloth composite	Non-woven cloth	Dip coating and polymerization	aqueous	H ₂ SO ₄	410 F/g at 0.5 A/g	26.6 Wh/kg	7000 W/kg	3000	90 %	[266]

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Table 5 (continued)

Electrode material	Flexible substrate	Fabrication process	Electrolyte type	Electrolyte material	Capacitance	Energy density	Power density	Cycling stability		Ref.
								Cycles	Capacitance retention (%)	
Carbon fiber yarn @carbon nanofibers @PANI nanowire	Carbon fiber yarn (CFY)	Electrospinning process and in situ polymerization	Polymer gel electrolyte	EMIMBF ₄ /PVDF/DMF	234 mF/cm ² at 0.1 mA/cm ²	21.4 μWh/cm ²	0.52 mW/cm ²	8000	90 %	[267]
Textile supercapacitors based on conducting polymers and other materials										
PANI/graphene/polyester textile	Polyester textile	In situ chemical polymerization	Polymer gel electrolyte	PVA/H ₂ SO ₄	1293 F/g at 1 A/g	N.R.	N.R.	3000	95 %	[268]
Cotton/graphene/PANI composite yarn	Cotton fibers	Coating and In-situ polymerization	aqueous	1 M H ₂ SO ₄	246 mF/cm ² at 5 mV/s	9.7 μWh/cm ²	840.9 μW/cm ²	3800	98 %	[269]
PANI/graphene/textile-HCl	Textile	Dip-and-dry process and in-situ polymerization	Polymer gel electrolyte	PVA/H ₂ SO ₄	1601 mF/cm ² at 1 mA/cm ²	755 mWh/m ²	1448 mW/m ²	10,000	75 %	[270]
PANI@GO coated silk fabric	Silk fabrics	In-situ polymerization and vacuum filtration	Polymer gel electrolyte	PVA/H ₃ PO ₄	71.2 F/g at 1 A/g	25.31 Wh/kg	8018 W/kg.	5000	87.4 %	[271]
PANI @ graphene-coated polyester textile (PANI/GPT)	Polyester textile (PT)	Electrochemical deposition and polymerization	Polymer gel electrolyte	PVA/H ₃ PO ₄	896.50 F/g at 1A/g	13.11 Wh/kg	3.00 kW/kg	5000	95.10 %	[272]
Carbon woven fabric/PANI/graphene composite	Carbon woven fabric	Electrodeposition and coating	aqueous	1 M H ₂ SO ₄	790 F/cm ² at 1 A/cm ²	28.21 uWh/cm ²	0.12 mW/cm ²	5000	88.9 %	[32]
Graphene/PANI-Coated Hierarchical Fabric	Multidimensional hierarchical fabric	Spraying method and electrochemical polymerization	Polymer gel electrolyte	PVA/PAM	707.9 mF/cm ² at 1 mA/cm ²	62.92 μWh/cm ²	0.39 mW/cm ²	10,000	80.8 %	[30]
PANI/RGO/PMFT flexible electrode	Polyester fiber textile	In situ polymerization and dip coating	Aqueous	1 M H ₂ SO ₄	564 mF/cm ² at 1 mA/cm ²	50.1 μWh/cm ²	20 mW/cm ²	10,000	94.4 %	[273]
PANI-PAAMPSA/PET	Polyethylene terephthalate (PET) fabric	Polymerization and coating	Aqueous	2 M H ₂ SO ₄	60 F/g at 20 mV/s	N.R.	N.R.	89 %	1000	[274]
PANI/AgNWs/cotton	Cotton fiber	In-situ polymerization	Aqueous	HCl	154 F/g at 0.5 A/g	N.R.	N.R.	96 %	5000	[275]
MoS ₂ /PANI/functionalized carbon cloth	Carbon cloth	A liquid-phase method, in situ polymerization, and drop-casting	Aqueous	1 M H ₂ SO ₄ (electrode) 1 M H ₂ SO ₄ (device)	452.25 F/g at 0.2 A/g 72.8 F/g at 0.2 A/g	N.R.	N.R.	1000	87 %	[276]
Textile supercapacitors based on conducting polymers and other materials										
NiCo ₂ O ₄ /PANI/carbon textiles composite	Carbon textiles	Hydrothermal and electrochemical deposition	Aqueous	6 M KOH	720.5C/g at 1 A/g	N.R.	N.R.	10,000	99.64 %	[277]
PANI-ZIF-67-CC	Carbon cloth (CC)	Dip coating and electrodeposition	Polymer gel electrolyte	PVA/H ₂ SO ₄ (electrode) PVA/H ₂ SO ₄ (device)	2146 mF/cm ² at 10 mV/s 35 mF/cm ² at 0.05 mA/cm ²	0.0161 mWh/cm ³	0.833 W/cm ³	2000	80 %	[278]
PANI/ZnO/ZIF-8/graphene/polyester textile	Polyester textile (PC)	Chemical and electrochemical deposition, coating	Polymer gel electrolyte	PVA/H ₂ SO ₄	1.378 F/cm ² at 1 mA/cm ²	235 μWh/cm ³	1542 μW/cm ³	400	73 %	[279]
Pt/CNT@PANI composite yarn.	CNT yarn	In situ polymerization and in situ deposition.	Polymer gel electrolyte	PVA/H ₃ PO ₄	91.67 mF/cm ² at 0.8 mA/cm ²	12.68 μWh/cm ²	399 μW/cm ²	5000	80 %	[280]
Pt/5-CNT@PANI FSSCs	Carbon nanotube yarn	In-situ polymerization	Polymer gel electrolyte	PVA-H ₃ PO ₄	217.7 F/g at 0.2 A/g	30.22 Wh/kg	91.88 W/kg	3000 5000	98.17 % 95.91 %	[281]

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Table 5 (continued)

Electrode material	Flexible substrate	Fabrication Process	Electrolyte type	Electrolyte material	Capacitance	Energy density	Power density	Cycling stability		Ref.
								Cycles	Capacitance retention (%)	
PET/Au/polyaniline hybrid	PET substrates	In situ electrodeposited	Polymer gel electrolyte	PVA/H ₂ SO ₄	51.7 mF/cm ² at 0.1 mA/cm ²	5.57 mW h/cm ³	0.33 W/cm ³	92.3 %	1000	[282]
PANI/CNT/Au/PET SC	PET fabric	Dip-coating and electroplating	Polymer gel electrolyte	PVA/H ₃ PO ₄	103 mF/cm ² at 1 mA/cm ²	241.2 μWh/cm ³	0.011 W/cm ³	89 %	2500	[283]
PANI/CNT/cellulose	Cellulose	In situ polymerization and coating	Aqueous	1 M H ₂ SO ₄	From 757 to 615 F/g at 5 mA/cm ²	N.R.	N.R.	81 %	1000	[284]
Hierarchical PANI/SnS ₂ @CNTs/carbon fibers	Carbon fibers	Hydrothermal, ICVD, coating, and in situ polymerization	Aqueous	1 M Na ₂ SO ₄	891 F/g at 20 mV/s	38.7 W h/kg	1 kW/kg	83.8 %	6000	[285]
NiCo ₂ S ₄ /PANI/CNTs hierarchical@GPAF	Graphene polyamide blend fiber (GPAF)	Hydrothermal strategy and in-situ polymerization	Polymer gel electrolyte	PVA-KOH gel	1290 mF/cm ² at 2 mA/cm ²	83.3 μWh/cm ²	420 μW/cm ²	80.13 %	5000	[286]
rGO/MnO ₂ /PANI/cotton fabric electrode	Textile cotton fabric	Dip coating, in-situ polymerization, and electrodeposition	Aqueous	1 M H ₂ SO ₄	888 F/g at 1 A/g	N.R.	N.R.	70 %	3000	[287]
RGO/cMWCNT//CFP/PPy	Carbon fiber paper	Vacuum-infiltration and electrochemical deposition method	Gel electrolyte	Potassium polyacrylate/KCl	82.4 F/g at 0.5 A/g	28.6 Wh/kg	15.1 kW/kg	93 %	2000	[288]

Electrode material	Flexible substrate	Fabrication process	Electrolyte type	Electrolyte material	Capacitance	Energy density	Power density	Cycling stability		Ref.
								Cycles	Capacitance retention (%)	
Textile supercapacitors based on conducting polymers and other materials										
PANI/CNTs/MnO ₂ /ACFC	Activated carbon fiber cloth (ACFC)	Electropolymerization process, dipping and drying method, and in situ chemical reaction.	aqueous	1 M H ₂ SO ₄ symmetric supercapacitor	4615 mF/cm ² at 5 mA/cm ²	157 μWh/cm ²	10,372 μW/cm ²	10,000	75 %	[289]
			Polymer gel electrolyte	PVA/H ₂ SO ₄ asymmetric supercapacitor		413 μWh/cm ²	16,120 μW/cm ²			
Graphene/CNTs hybrids/PANI composite fiber	Nickel textile	CVD method and electrochemical deposition	Polymer gel electrolyte	PVA/H ₃ PO ₄	164 mF/cm ²	9.196 μWh/cm ²	0.28 mW/cm ²	4500	94 %	[290]
Lignosulfonate/PANI/functionalized graphene hydrogel (FGH)/FCC	Carbon cloth	Hydrothermal process, chemical deposition and in situ polymerization	aqueous	1 M H ₂ SO ₄	1223 mF/cm ² at 2 mA/cm ²	160.6 μWh/cm ²	1000 μW/cm ²	5000	86 %	[291]
Hyaluronic acid (HA), carbon nanotubes (CNTs), polyaniline (HA/CNT/PANI fibers)	Fiber	Wet spinning and electrochemical polymerization.	aqueous	0.5 M H ₂ SO ₄	282.36 ± 90.93 mF/cm ²	N.R.	N.R.	3000	90 %	[292]
Nitrogen-doped graphene/polyacrylic acid/polyaniline composites	Carbon cloth	In-situ polymerization and coating	Solid gel electrolyte	H ₂ SO ₄ -PVA (electrode) H ₂ SO ₄ -PVA (device)	521 F/g at 0.5 A/g 68 F/g at 1A/g	5.8 Wh/kg	1.1 kW/kg	2000	83.2 %	[293]
PPy/GO/PET fabric	PET non-woven fabrics	In-situ polymerization	aqueous	2.0 M NaCl	568.35 F/g at 0.1 A/g	37.4 Wh/kg	N.R.	500	83.4 %	[294]
PPy/RGO/cotton	Cotton fabric	Thermal reduction of GO and chemical polymerization	aqueous	2.0 M NaCl	336 F/g at 0.6 mA/cm ²	21.1 Wh/kg	N.R.	500	64 %	[295]
PPy/rGO nanocomposite cotton fabric (NCF)	Cotton fabric	Chemical polymerization	Polymer gel electrolyte	PVA/H ₂ SO ₄	9300 mF/cm ² at 1 mA/cm ²	167 μWh/cm ²	1.20 mW/cm ²	10,000	94.47 %	[296]
Reduced graphene oxide/	Polyethylene terephthalate (PET) fabric	Dip coating and polymerization	Polymer gel electrolyte	PVA-H ₂ SO ₄	0.23 F/cm ² at 1 mV/s	11 μWh/cm ²	0.03 mW/cm ²	6000	76 %	[43]

(continued on next page)

Table 5 (continued)

Electrode material	Flexible substrate	Fabrication process	Electrolyte type	Electrolyte material	Capacitance	Energy density	Power density	Cycling stability		Ref.
								Cycles	Capacitance retention (%)	
polypyrrole composite PET/rGO-x/PPy										
Electrode material	Flexible substrate	Fabrication process	Electrolyte type	Electrolyte material	Capacitance	Energy density	Power density	Cycling stability	Capacitance retention (%)	Ref.
Textile supercapacitors based on conducting polymers and other materials										
Cotton/PPy/MWCNT composite	Cotton textile	In situ chemical deposition method.	Polymer gel electrolyte	(PVA)/H ₃ PO ₄	206.8 F/g at 1 mA/cm ²	19.6 Wh/kg	201 W/kg	400	72 %	[297]
MWCNT/PPy/cotton	Cotton yarn	Polymerization process and coating	Polymer gel electrolyte	PVA)/H ₃ PO ₄	30 F/g	2.63 mW h/g	11.33 mW/g	N.R.	N.R.	[298]
PPy/MWCNT/cotton fabric	Cotton	Electrodeposition and electrochemical polymerization	Polymer gel electrolyte	H ₃ PO ₄ /PVA	201.99 F/g at 1.8 A/g	64.64 W h/kg	5.14 kW/kg	375	93 %	[299]
PPy@CNTs@urethane elastic fiber core yarn	Urethane elastic fiber	Dipping and electrodeposition	Polymer gel electrolyte	PVA/H ₃ PO ₄	69 mF/cm ² at 5 mV/s	0.00613 mW h/cm ²	0.133 mW/cm ²	N.R.	N.R.	[300]
PPy coated carbonized silk fabric	Silk fiber	Electro-deposition method	Polymer gel electrolyte	PVA/H ₃ PO ₄ (electrode)	4040 mF/cm ² at 2 mA/cm ²	6.88 mWh/cm ³	0.04 W/cm ³	1500	88.6 %	[301]
				PVA/H ₃ PO ₄ (device)	666.78 mF/cm ² at 2 mA cm ⁻²					
PPy/bacterial cellulose (BC)/Cotton Yarn	Cotton yarns	Coating and deposition	Polymer gel electrolyte	PVA/H ₂ SO ₄	76.6 m F/cm ² at 0.42 mA/cm ²	16.9 μWh/cm ²	10.9 μW/cm ²	N.R.	N.R.	[302]
PPy-based hybrid nanostructures (VPPyNTs/PPyG-textile)	Textile substrate	Polymerization process	Polymer gel electrolyte	PVA/H ₃ PO ₄	64 F/g	N.R.	N.R.	500	99 %	[303]
EPPy-PPy/NF/CF	Cotton fabric (CF)	Electropolymerization and spray Coating	Polymer gel electrolyte	PVA/H ₂ SO ₄	669 mF/cm ² at 0.2 mA/cm ²	59.5 μWh/cm ²	0.04 mW/cm ²	10,000	100 %	[304]
CPYF-ZIF-67-PPy	Polymer yarns.	CVD process, and in-situ polymerization	Aqueous	0.5 M H ₂ SO ₄	2308.8 mF/cm ² at 0.5 mA/cm ²	112 μWh/cm ²	201.5 μW/cm ²	10,000	87.6 %	[305]
PPy/TiO ₂ -coated cotton fabric	Cotton fabric	Sol-gel method and chemical oxidation.	Aqueous	2.0 M NaCl	733 F/g at 0.6 A/cm ²	44.4 Wh/kg	555 W/kg	N.R.	N.R.	[306]
Polypyrrole-coated manganese dioxide CC/MnO ₂ /PPy//Co ₃ O ₄ /CC	Carbon cloth	CVD, dip coating and polymerization	Polymer gel electrolyte	PVA/KOH	59.5 F/cm ³ at 20 mA/cm ²	27 mWh/cm ³	1.31 W/cm ³	5000	96 %	[307]
Electrode material	Flexible substrate	Fabrication process	Electrolyte type	Electrolyte material	Capacitance	Energy density	Power density	Cycling stability	Capacitance retention (%)	Ref.
Textile supercapacitors based on conducting polymers and other materials										
PPy/MnO ₂ on carbon cloth	Carbon cloth	In-situ electrodeposition	aqueous	1.0 M Na ₂ SO ₄	325 F/g at 0.2 A/g	N.R.	N.R.	1000	96 %	[308]
PPy/MoS ₂ /CC	Carbon cloth	Hydrothermal and electrochemical deposition.	aqueous	5 M LiCl Electrode	1150.4 mF/cm ² at 1 mA/cm ²	1.412 mWh/cm ³	10 mW/cm ³	5000	87.2 %	[309]
				5 M LiCl Device	1016.4 mF/cm ² at 2 mA/cm ²					
CF/MnO ₂ /PPy	Carbon fiber	Electrodeposition and dip coating	Polymer gel electrolyte	PVA/H ₃ PO ₄	69.3 F/cm ³ at 0.1 A/cm ³	6.16 mWh/cm ³	0.04 W/cm ³	1000	86.7 %	[310]
Polypyrrole-coated stainless steel/cotton (PPy/SS/cotton)	Cotton	Electrochemical polymerization process and coating	Polymer gel electrolyte	PVA/H ₂ SO ₄	344 mF/cm ² at 0.6 mA/cm ²	36.2 μWh/cm ²	135 μW/cm ²	1000	93 %	[49]
CNT/rGO and PPy coated cotton	Cotton	Dip-coating and chemical polymerization,	aqueous	1 M Na ₂ SO ₄	569.6 mF/cm ² at 1 mA/cm ²	0.26 mW h/cm ² ,	N.R.	1000	91 %	[311]

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Table 5 (continued)

Electrode material	Flexible substrate	Fabrication process	Electrolyte type	Electrolyte material	Capacitance	Energy density	Power density	Cycling stability		Ref.	
								Cycles	Capacitance retention (%)		
Polypyrrole/Ag/graphene oxide nanocomposite	Cotton fabric	UV-induced polymerization	Polymer gel electrolyte	PVA/H ₂ SO ₄	286.6 mF/cm ² (4.7 F/cm ³) at 0.5 mA cm ⁻²	25.5 μWh/cm ²	1149.5 μW/cm ²	10,000	89.7	[312]	
Polypyrrole – MnO ₂ - Coated CNT - cotton (PMCCT)	CNT textile (cotton)	Dip Coating and electrochemical deposition	aqueous	1 M Na ₂ SO ₄	461 F/g at 0.2 A/g.	31.1 Wh/kg	22.1 kW/kg	10,000	93.8 %	[313]	
PPy@MnO ₂ @rGO@conductive yarns.	Stainless steel fiber yarn	Hydrothermal, electrodeposition and polymerization	solid-state electrolyte	PVA/H ₃ PO ₄	411 m F/cm ² 31 mF/cm at 11 mA/cm ³	0.0092 mWh/cm ² , 1.1 mWh/cm ³	1.33 mW/cm ² , 160 mW/cm ³	4950	92 %	[314]	
PPy-MnO ₂ -SWCNTs-cotton thread electrodes	Cotton thread	Electrochemical deposition and polymerization	Aqueous	Na ₂ SO ₄	36.6 mF/cm 486 mF/cm ²	1.49 F/cm ² at 1 mV/s	33 μ Wh/cm ² 14.7μWh/cm ²	0.67 mW/cm ² 13 mW/cm ²	2000	87 %	[315]
PPy/rGO/SnCl ₂ modified polyester yarn	Polyester fiber	Electrochemical deposition	Polymer gel electrolyte	PVA/H ₂ SO ₄	474 mF/cm ² at 1 mA/cm ²	0.0658 mWh/cm ²	25 mW/cm ²	10,000	100 %	[316]	

Electrode material	Flexible substrate	Fabrication process	Electrolyte type	Electrolyte material	Capacitance	Energy density	Power density	Cycling stability		Ref.
								Cycles	Capacitance retention (%)	
Textile supercapacitors based on conducting polymers and other materials										
RGO/MoS ₂ /PEDOT@carbon fabric	Carbon fiber cloth	Hydrothermal, polymerization	Aqueous	1 M H ₂ SO ₄	241.81 mF/cm ² 0.5 mA/cm ²	1.44 μWh/cm ²	0.058 mW/cm ²	5000	93.7 %	[317]
MnO ₂ /PEDOT:PSS-rGO@carbon fiber	Carbon fiber	Drop coating and electrodeposition	Aqueous	1 M Na ₂ SO ₄	2.9 F/cm ² (194 F/cm ³ , 550 mF/cm) at 5 mA/cm ²	295 μWh/cm ² 19 mWh/cm ³	2900 μW/cm ² 190 mW/cm ³	5000	95 %	[318]
Chitosan/MnO ₂ @MnCO ₃ chelate decorated fabrics	Carbon fabrics	Hydrothermal method	Ionic liquid electrolyte	BMIMBF ₄	2.5 F/cm ² at 0.25 mA/cm ²	0.35 Wh/cm ²	N.R.	10,000	~90 %	[319]
PEDOT:TREN:MnO2@MnCO3	Textile	Hydrothermal method	Sweat electrolyte	sweat	30 F/cm2 at 0.25 mA/cm ²	N.R.	4.16 Wh/cm ²	50,000	92 %	[320]
MnO ₂ -CNT/CNT PEDOT:PSS ink/nylon/spandex fabric	Nylon/spandex fabric	Screen-printing method	Gel electrolyte	1 M LiCl-PVA	100mF/cm ² at 0.24 mA/cm ²	17.5 μW/cm ²	0.4 mW cm ²	N.R.	N.R.	[321]
MOF-6/RGO/PPy@PET fabric	PET fabric	Dip coating, reduction and polymerization	Polymer gel electrolyte	PVA/H ₂ SO ₄	3.5 F/cm ³ 90 mF/cm2 at 5 mV/s	64 μW h cm ⁻³	0.6 mW cm ⁻³	1000	85 %	[322]
GNS/PEDOT:PPS-coated hierarchical fabric	Hierarchical fabric	Spraying and screen-printing method	Polymer gel electrolyte	PVA/H ₂ SO ₄	245.5 mF/cm ² at 1 mV/cm ²	21.82 μWh/cm ²	0.4 mW/cm ²	10,000	95 %	[31]
CC/Ti-Fe ₂ O ₃ @PEDOT//MnO ₂ /CC	Carbon cloth	Hydrothermal, electrodeposition	Polymer gel electrolyte	PVA/LiCl	2.4 F/cm ³ at 1 mA/cm ²	0.89 mWh/cm ³	440 mW/cm ³	6000	85.4 %	[323]
rGO NSs/polyamide-66 nanofiber fabric	Polyamide textile	Dipping process (electrospinning)	Aqueous	1 M H ₂ SO ₄	931 mF/cm ² 38.79 F/cm ³ at 0.5 A/g	7.6 Wh/kg	1500 W/kg	N.R.	N.R.	[324]
Nitrogen-doped carbon derived from polyimide/MWCNT composites (C-MWCNT-PI/CC)	Carbon cloth	Carbonization, polymerization, and coating	Polymer gel electrolyte	PVA/H ₂ SO ₄	3.88 F/cm ³ at 0.02 mA/cm ³	0.50 mWh/cm ³	72 mW/cm ³	10,000	85.3 %	[325]

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Table 5 (continued)

Electrode material	Flexible substrate	Fabrication Process	Electrolyte type	Electrolyte material	Capacitance	Energy density	Power density	Cycling stability		Ref.
								Cycles	Capacitance retention (%)	
Textile supercapacitors based on carbon materials and other materials										
MWCNT/RGO nanocomposite/textile	Spandex/nylon textile	Dip-coating	Gel-type electrolyte	PC-PMMA-[BMIM][TFSI]	30.4 mF/cm ² at 0.025 mA/cm ²	4.63 μWh/cm ²	24.7 μW/cm ²	10,000	99.7 %	[72]
MWCNT/MoO ₃ nanocomposite in the fabric	Nylon fabric	Spray-coating	Gel-type electrolyte	ACN – PC–PMMA–LiClO ₄	48.3 F/g at 0.14 A/g	13.15 Wh/kg	100 W/kg	5000 10,000	86 % 76 %	[326]
CNPs/rGO-CT	Cotton threads (CT)	Dip coating and low-temperature vapor reduction method	Polymer gel electrolyte	PVA-H ₃ PO ₄	3.79 m F/cm ³ at 50 mV/s	0.084 μWh/cm ³	35.3 μW/cm ³	10,000	95.23 %	[327]
AgNPs/RGO/CF composite	Cotton fabric (CF)	Electroless silver plating and dip coating	Aqueous	0.5 M NaOH	426 ± 10 F/g at 5 mV/s	34.6 Wh/kg	125 W/kg	1000	89 %	[328]
RGO/Ni cotton yarn composite	Cotton yarns	Electroless and electrochemical deposition	Polymer gel electrolyte	(PVA)/LiCl	292.3 F/cm ³ at 87.9 mA/cm ³ 0.11 F/cm	6.1 mWh/cm ³	1400 mW/cm ³	10,000	82 %	[329]
RGO muffled cobalt oxide on flexible carbon fabric (CONW-RGO)	Carbon fabric substrate	Hydrothermal method	Polymer gel electrolyte	PVA/KOH	1110 F/g at 1 A/g	34.78 Wh/kg (0.214 mWh/cm ³)	3.6 kW/kg (23 mW/cm ³)	10,000	86.4 %	[330]
Graphene/MnO ₂ /cellulose nonwoven fabric (GMNF-SC).	Cellulose nonwoven fabric	Coating and in-situ chemical growth method	Polymer gel electrolyte	H ₂ SO ₄ /PVA	138.8 mF/cm ² at 0.5 mA/cm ²	12.34 μWh/cm ²	200 μW/cm ²	5000	82.5 %	[331]
RGO/Ni-MOF on Ni-coated polyester fabric	Polyester fabric	Hydrothermal and Dip-coating	Polymer gel electrolyte	1 M KOH/PVA	260 mF/cm ² at 4 mA/cm ²	7.72 μWh/cm ²	3.07 mW/cm ²	1500	70 %	[332]
Porous WO ₃ /graphene/polyester textile (WO ₃ /G/PT)	Polyester textile	Dip coating and electrodeposition	Gel electrolyte	PVA-H ₂ SO ₄	308.2 mF/cm ² 167.6 mF/cm ³ at 2 mV/s	60 μWh/cm ³	2320 μW/cm ³	N.R.	N.R.	[333]
RGO and MnO ₂ /m-CC	Porous carbon cloth	Dip coating and microwave irradiation	Polymer gel electrolyte	PVA/Na ₂ SO ₄	264 and 331 mF/cm ² at 1 mA/cm ²	64 μW h/cm ²	1 mW/cm ²	10,000	95 %	[334]
MnO ₂ and ErGO/SPNFs	Silver-plated nylon fibers (SPNFs)	Electrodeposition	Polymer gel electrolyte	PVA/Na ₂ SO ₄	1.02 F/cm ² (36.59 F/g) at 7.5 mA/cm ² (0.275 A/g)	1.02 mWh/cm ² (9.96 Wh/kg)	5.27 mW/cm ² (0.19 kW/k)	5000	85.2 %	[335]

Electrode material	Flexible substrate	Fabrication process	Electrolyte type	Electrolyte material	Capacitance	Energy density	Power density	Cycling stability		Ref.
								Cycles	Capacitance retention (%)	
Textile supercapacitors based on carbon materials and other materials										
RGO/MoO ₂ /carbon textile	Carbon textile	Solvothermal method and coating	aqueous	1 M H ₂ SO ₄	8132 mF/cm ² at 2 mV/s	143 μWh/cm ²	2176 μW/cm ²	30,000	95 %	[336]
GaN NW/graphite paper nanocomposites	Graphite paper	CVD method	Polymer gel electrolyte	H ₂ SO ₄ /PVA	237 mF/cm ² at 0.1 mA/cm ²	0.30 mWh/cm ³	1000 mW/cm ³	10,000	98 %	[337]
Graphene/MnO ₂ -textile as positive and SWNTs-textile as negative electrode	Polyester fabric	Coating	aqueous	Na ₂ SO ₄	up to 315 F/g	12.5 Wh/kg	110 kW/kg	5000	95 %	[338]
MnO ₂ /rGO@carbonized cotton fabrics	Carbonized cotton fabrics	Dip coating, chemical deposition, and carbonization	solid-electrolyte	1 M LiPF ₆ -EC-DMC-DEC	329.4 mA h/g at 100 mA/g	N.R.	N.R.	70	93.7 %	[339]
MnO ₂ /rGO@C	Polyester fabric	Hydrothermal method.	aqueous	1 M Na ₂ SO ₄	332 F/g at 2 mV/s	N.R.	N.R.	1000	100 %	[340]
MnO ₂ -CNT-textile	Polyester fabric	Dip coating	aqueous	0.5 M Na ₂ SO ₄	410 F/g at 5 mV/s	≈5–20 Wh/kg	13,000 W/kg	2000 10,000 50,000	80 % 60 % 50 %	[341]

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Table 5 (continued)

Electrode material	Flexible substrate	Fabrication process	Electrolyte type	Electrolyte material	Capacitance	Energy density	Power density	Cycling stability		Ref.
								Cycles	Capacitance retention (%)	
Two coiled MnO ₂ /CNT/nylon fiber electrodes	Nylon fiber	Electrochemical deposition	Polymer gel electrolyte	PVA-LiCl	5.4 mF/cm, 40.9 mF/cm ² at 10 mV/s	2.6 μWh/cm ²	66.9 μW/cm ²	N.R.	N.R.	[11]
ACFC/MnO ₂ /CNTs composites.	Activated carbon fiber cloth (ACFC)	Dip coating and electro-deposition	aqueous	KOH	2542 mF/cm ² at 2 mV/s	56.9 μWh/cm ²	16,287 μW/cm ²	1500	96 %	[342]
MnO ₂ -coated SWNT/Cotton fiber	Cotton fiber	Dipping and drying process	Aqueous	2 M Li ₂ SO ₄	0.48 F/cm ² , 140 F/g at 20 μA/cm ²	20 Wh/kg	10 kW/kg	8000 130,000	94 %	[343]
MnO ₂ -NiCo ₂ O ₄ //rGO	Bamboo fabric	Inkjet printing technique	Polymer gel electrolyte	LiCl/PVA	2.12 F/cm ² (1766 F/g) at 2 mA/cm ²	37.8 mWh/cm ³	2678.4 mW/cm ³	5000	93.1 %	[37]
Ag Np/activated carbon	Polyester fabric	Spray-coating technique	Polymer gel electrolyte	PVA/H ₂ SO ₄	25.7 F/g at 0.1 A/g	64.3 Wh/kg	1.5 Wh/kg	1000	25 %	[344]
RuO ₂ /SWCNT/cloth fabric	Cloth fabric	Inkjet-printing method	Polymer gel electrolyte	PVA/H ₃ PO ₄	138 F/g	18.8 Wh/kg	96 kW/kg	1000	~100 %	[345]

high ionic conductivity, low internal resistance, and low cost and are widely available compared to ionic liquid and organic electrolytes [358]. However, the main drawbacks of aqueous electrolytes are their narrow ESPW, the potential for leakage in flexible textiles and the limited operating temperature range of TSCs with aqueous electrolytes (above the water freezing point and below the boiling point). Another drawback when using a concentrated electrolyte is the corrosion at the electrode substrate surface, which may lead to the electrode material delaminating from the substrate [346,359]. Therefore, optimizing the electrolyte concentration with respect to the overall performance of the electrochemical supercapacitors is necessary. Neutral aqueous electrolytes have also been widely studied for supercapacitors due to larger working potential windows. Additionally, neutral solutions are not

corrosive and are intrinsically safer. However, the ionic conductivities and ESRs of SCs using neutral electrolytes are generally lower than those using the H₂SO₄ or the KOH electrolyte [360–362].

Organic electrolytes for supercapacitors typically consist of salts dissolved in organic solvents. Organic electrolytes dominate the commercial market due to their much wider voltage window of about 3 V. However, some issues should be considered when organic electrolytes are used for supercapacitors. Supercapacitors with organic electrolytes usually have a higher cost, a smaller specific capacitance, and a lower ionic conductivity. Safety concerns are related to the flammability, volatility, and toxicity compared to supercapacitors using aqueous electrolytes. Furthermore, an organic electrolyte salt requires strict purification and preparation procedures to obtain pure electrolytes and to

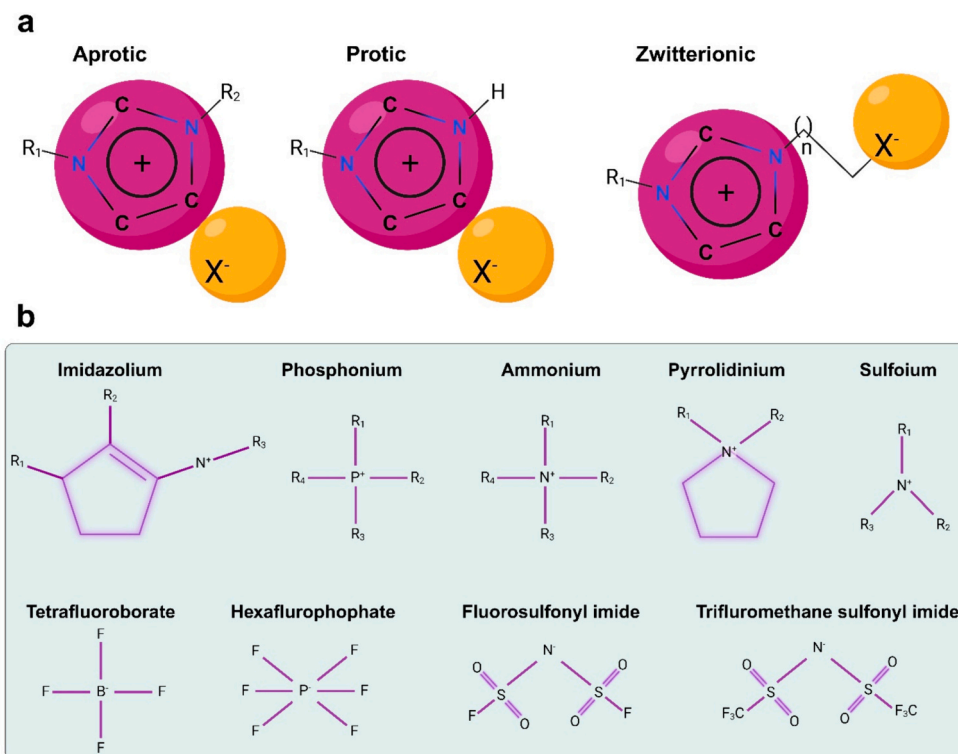


Fig. 4. a) From left to right shows aprotic, protic, and zwitterionic liquid electrolyte molecules, where for aprotic ILs electrolytes a diazolic ring is attached to two different functional groups and has a negative charge, protic IL electrolytes feature a diazolic ring with a hydrogen termination and is connected to one functional group and has a negative charge, and in zwitterionic IL, the diazolic ring is attached to a functional group and a negatively charged termination connected via a carbon chain; b) commonly encountered ionic groups used for preparing molten salts used in electrochemical studies.

remove any residual impurities (e.g., water) that can lead to large performance degradation and serious self-discharge issues [346,350]. Propylene carbonate is the most commonly used solvent in organic electrolytes due to its environmentally friendly nature, wide voltage window, and sufficient ionic conductivity [363].

Due to their distinct ionic properties and low volatilities, ionic liquid (IL) electrolytes have received significant interest as alternative electrolytes for supercapacitors. ILs are salts containing ions (cations and anions) with melting points below 100 °C [358,364]. They can be classified as aprotic, protic, and zwitterionic types based on their molecular nature. Fig. 4a presents some of the frequently encountered ILs used as electrolyte components. All three types of ILs feature a diazole ring. For aprotic ILs, the diazolic ring is connected to two different functional groups and has a negative charge. For protic ILs, the negatively charged diazolic ring is connected to one functional group and features one hydrogen surface termination. For zwitterionic ILs, the neutral diazolic ring is connected to a functional group and a carbon chain that has a negatively charged termination. Aprotic ILs are suitable for energy storage devices (supercapacitors and lithium batteries) [346,350]. In the literature, cations of IL electrolytes commonly used for constructing supercapacitors include imidazolium, phosphonium, ammonium, pyrrolidinium, and sulfonium. Anions commonly used in IL electrolytes include tetrafluoroborate $[\text{BF}_4]^-$, hexafluorophosphate $[\text{PF}_6]^-$, Bis(fluorosulfonyl imide) $[\text{FSI}]^-$, and Bis(Trifluoromethane sulfonyl imide) $[\text{TFSI}]^-$ all shown in Fig. 4b. In general, the imidazolium-based ILs can provide higher ionic conductivity, while the pyrrolidinium-based ILs have larger ESPWs [358,365]. IL electrolytes have several potential advantages, including a wide voltage window, high thermal and chemical stability, negligible volatility, and non-flammability (depending on the combination of cations and anions) [346,350,366]. The physical and chemical properties of IL electrolytes are tunable as anions can be made task-specific [367]. Therefore, IL electrolytes can overcome many disadvantages of the conventional aqueous and organic electrolytes. Unfortunately, there are several drawbacks with most ionic liquid electrolytes, such as high viscosity, low ionic conductivity, and high cost, which can limit their practical use in supercapacitors [346,350,366].

Even though liquid electrolytes demonstrate desirable conductivity, there are several drawbacks to their use in wearable supercapacitors, such as fire hazards, low cycling stability, leakage problems, and toxicity to humans when incorporated into wearable devices [146,368–371]. The supercapacitors need to be thoroughly and constantly soaked in the electrolyte to work properly; the optimal conditions can be created and maintained in a lab setting but are less feasible during practical processes. Creative approaches such as using human sweat as an electrolyte have been pursued; however, the performance of sweat-based TSCs, when tested under practical conditions, was far from performance under laboratory conditions [146]. In addition to these problems, there are other practical barriers: nobody wants to wear wet clothing, and fewer still want to wear clothing that must be constantly soaked in sweat to charge their devices. While these devices could be surrounded in a flexible casing, the issues associated with electrolyte leakage and durability will remain [372]. The development of gel and solid electrolytes allows for solving many of the problems posed by liquid electrolytes in wearable supercapacitors [368,370].

4.2. Solid- or quasi-solid-state electrolytes

Solid or gel electrolytes have attracted great interest in recent years for flexible and wearable supercapacitors. The solid-state electrolytes can function simultaneously as the ionic conducting media and as a separator [346]. The main advantage of using solid or quasi-solid-state electrolytes is avoiding the potential leakage problem of liquid electrolytes [373,374]. Solid-state electrolytes are based on oxides, sulfides, carbides, or polymers [375]. Given the TSCs' flexibility requirement, most research in TSCs has been focused on soft and flexible materials.

This need has directed research into solid polymer electrolytes (SPEs), also called solid electrolytes, and quasi-solid-state electrolytes (QSSE), or ionogels.

Solid electrolytes are composed of a polymer (e.g., PEO) and a salt (e.g., LiCl) without any solvents (e.g., water). The ionic conductivity of solid electrolytes is provided by the transportation of salt ions through the polymer [376]. Solid electrolytes are more stable, have a wider electrochemical voltage window, and have a longer lifetime than liquid electrolytes [368]. Solid electrolytes can also be redissolved and reused with negligible performance change, so devices utilizing them could require fewer resources in their fabrication and be easier to recycle [377]. Polyvinyl alcohol (PVA) and poly (methyl methacrylate) (PMMA) have been used for incorporation into TSCs due to their stability, transparency, non-toxicity, and good ionic conductivity [7,72]. Natural plasticizer sources, such as gelatin, are being explored as more sustainable alternatives for solid electrolytes [378].

Gel electrolytes, or QSSE, consist of a polymer host and a liquid electrolyte (e.g., organic electrolyte and ILs electrolyte) [346]. The ionic conductivity of gel electrolytes is much higher than that of solid electrolytes due to a liquid phase in the gel electrolytes. They have attracted attention in research because gel electrolytes combine the high ionic conductivity of liquid-based electrolytes with the high stability of solid-based electrolytes. The ionic conductivity of the gel electrolyte is provided by the transport of salt ions through the solvent and not through the polymer [374]. It has been observed that gel electrolytes improve supercapacitor performance due to the slow permeation of gel throughout the supercapacitor [7]. Therefore, gel electrolytes are currently the most extensively studied electrolytes in wearable supercapacitors. However, identified issues, such as relatively poor mechanical strength and narrow operating temperature range, especially when using water as a solvent, must be considered when using gel electrolytes in wearable supercapacitors [379]. The displacement of the liquid phase under mechanical strength should be another avenue research must address when using gel electrolytes.

Hydrogel electrolytes are formed using a polymer and water as dispersing medium and behave as a solid due to the electrostatic forces holding the polymer structure together [371,374,380]. They can suffer from limited operating cell voltages and thus low energy density due to the narrow ESPW of the aqueous component in the hydrogel. The gel electrolyte is made of a polymer with low ionic conductivity, so it is combined with an ionic compound such as sulfuric acid, phosphoric acid, potassium hydroxide, potassium ferricyanide, potassium chloride, or sodium chloride to improve the ionic conductivity and the electrochemical performance of textile supercapacitors [381]. IL gel polymer electrolytes have attracted wide interest in electrochemical energy storage devices, particularly for their use in flexible supercapacitors [381,382]. IL gel electrolytes are made by incorporating IL into polymer hosts [383]. The main advantages of IL gel electrolytes are a wide operating potential range, high ionic conductivity, and high stability. These advantages depend on the nature of the IL and the interaction between the host polymer [381–385].

Balancing air permeability and performance in textile supercapacitors is a critical challenge, especially when considering the use of gel electrolytes, which can compromise permeability and comfort. Some strategies to achieve this balance include optimizing the electrolyte composition, advanced fabrication and encapsulation techniques, material innovation, and structural substrate design. Using ionic liquid-based gel electrolytes [383] or gel electrolytes doped with nanoparticles or other conductive materials can provide high ionic conductivity with improved mechanical properties and flexibility, allowing for thinner and more porous layers that do not significantly impair air permeability. Creating breathable assembly for the electrolytes and electrodes can be envisioned as another avenue for research. For instance, the electrospinning technique can be used to tune fibers that remain air permeable upon coating with gel electrolyte [233,386–388]. There are reports on using a layer-by-layer technique to layer electrodes

and electrolytes in a series of concentric shells in one fiber, controlling the thickness and distribution of each layer, including the gel electrolyte, and allowing for optimizing both the electrical performance and air permeability of the textile [389–392]. Using breathable but water-resistant encapsulation materials can further protect the supercapacitor while allowing moisture and air to move through the textile to maintain comfort and air permeability [393–395]. Similarly, using textiles with inherent porosity or designing multi-layer textiles with integrated air channels can help maintain air permeability while embedding the supercapacitor elements [396]. Conductive polymers like poly(3,4-ethylenedioxythiophene) (PEDOT) and hybrid materials utilizing graphene can be used to create breathable electrodes that require less gel electrolyte [397–401]. By combining these strategies, it is possible to develop textile supercapacitors that balance air permeability, comfort, and electrochemical performance, making them viable for practical applications in wearable electronics.

5. Smart textiles market and applications of textile supercapacitors

In recent years, intelligent and soft wearable textiles have shown tremendous potential to replace rigid wearable electronics systems. The global smart textiles market may reach ~\$11.5 billion by 2027, as evidenced by research and market reports, and it is expected that the size of the smart textiles market will exceed 32.3 % of the global textile market [402–404]. Hence, the versatility of design, flexibility, and compact nature of TSCs hold great promise for their incorporation into clothing, unlike the current rigid capacitors and batteries that are standard components of existing electronics [405–407]. The potential applications of smart, flexible, and wearable textile supercapacitors in medicine, military, sports, and consumer sectors are being explored. Biomonitoring in medicine, sports, and consumer markets is a method of health monitoring growing in popularity, being a well-established standard of care within medical settings, and soon becoming common in sports settings [47]. Fig. 5 shows potential areas of application for wearable supercapacitors.

5.1. Medical applications

In medical applications, flexible and wearable textile supercapacitors [37,47,52,408] have potential uses in monitoring bio-signals,

temperature, and motion and supporting non-invasive diagnostic tests, as shown in Fig. 5. Sensing bio-signals with wearable textiles for continuous healthcare monitoring is currently gaining attention [409,410]. Additionally, flexible supercapacitors have been studied for their possible applications as a power source for medical implant devices, such as heart and glucose monitors [411,412]. These devices are more compact and flexible than conventional batteries used in standard medical implants, increasing the user's comfort. When flexible supercapacitors are combined as one component of an overall circuit designed to collect and store energy for an implanted medical device, they address a significant challenge: the need for expensive and potentially risky surgeries to replace worn-out batteries [354]. When used in sports applications, textile supercapacitors can monitor physiological responses when athletes are at the gym or in a match, providing the person exercising or the medical officer with the needed information about the performance of the athletes [19,409]. TSCs can also be used to create self-powered pulse monitors; Meng et al. created a silver-coated fabric with wireless communication that could be placed over pulse points for biomonitoring that was successfully used in diagnosing sleep apnea-hypopnea syndrome [408].

5.2. Military applications

In military applications, the incorporation of flexible supercapacitors into smart clothing that can collect and store energy from body heat and movement is being explored [406]. The goal is to replace battery packs currently used by soldiers susceptible to bullet puncture and crushing. Additionally, this approach will reduce the load of the soldiers' packs [413,414]. E-textiles can also be used as protective garments by providing heating or cooling to the user in extreme environments or alerting the user to changes in bio-signals [415]. Smart textiles in defense can be used to monitor soldiers' physiological condition, and antennas can be incorporated into a textile-based patch and attached to soldiers' protective wear for close-range communication and coordination to reduce casualties [416,417]. Additionally, flexible supercapacitors are resilient enough to provide a reliable power source after being physically damaged, unlike rechargeable batteries that cease functioning, are combustible, and must be discarded [418]. Resilient materials that continue functioning after use and abuse are essential components of protective garments worn by soldiers on military expeditions due to the harsh environments and rough treatment to which

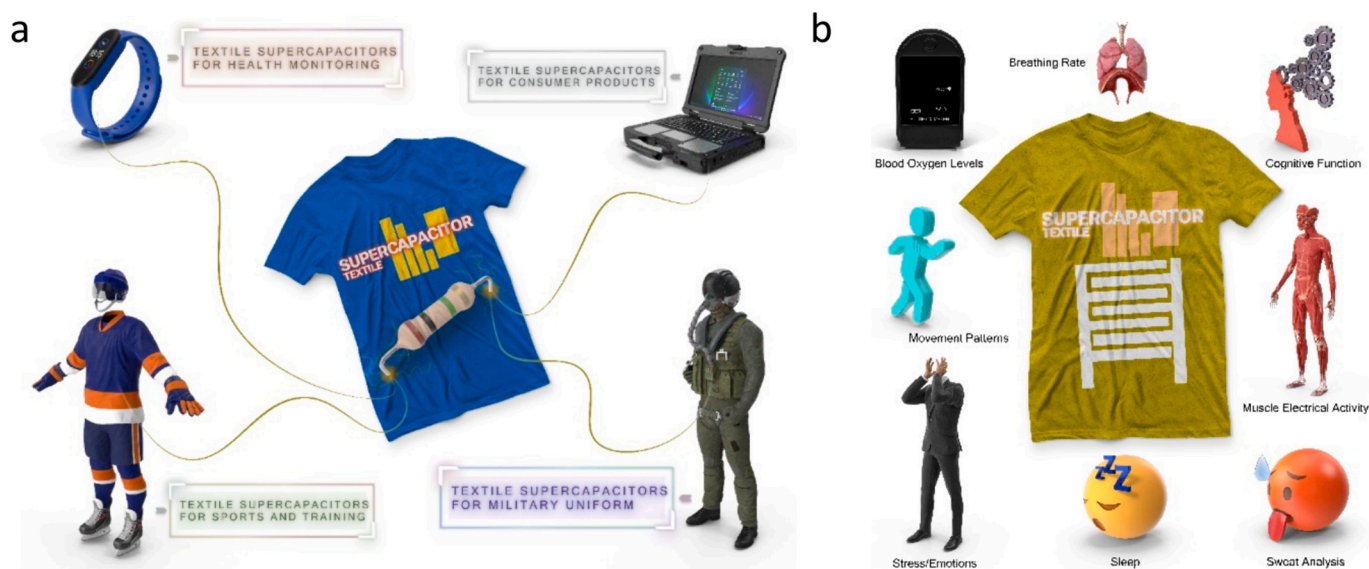


Fig. 5. Applications for flexible supercapacitors (a) in health, military, sports, and consumer markets, (b) showing the possible applications of TSCs to monitor breathing rate, cognitive function, muscle activity, sweat, recharge medical devices, stress, and sleep.

these textiles are often subjected [419]. Researchers have also explored creating self-powered equipment by integrating TSCs into a braided rope to power emergency signals for rescue [420]. It is expected that in the future, the lessons learned from military use of energy-storing textiles will lead to their utilization by the general populace, and they will become a critical component of rescue and exploration missions.

6. Life cycle assessment of textile supercapacitors

As the demand for energy storage devices grows, it is essential to consider the environmental impacts associated with the materials used in TSCs. This section aims to explore the sustainability aspects of the materials employed in textile-based supercapacitors via their life cycle assessment (LCA) and environmental impact, including electrode materials such as carbon-based nanomaterials, conductive polymers, and MXenes. Additionally, the choice of substrate materials and electrolytes is examined in terms of their environmental impact. Potential strategies to mitigate their carbon footprint will be discussed.

LCA is becoming a common technique for analyzing the burden that a technology's entire life cycle poses on many facets of the environment. The 'Cradle-to-Grave' approach considers a new technology's production, use, and eventual end-of-life phases. LCA compares the environmental impacts to known processes to guide future research and innovation toward the most sustainable and environmentally friendly materials, processes, distribution, and eventual recovery. LCA software and expansive up-to-date LCA databases, such as OpenLCA, assist researchers in identifying and comparing their new technology to older technologies in many classifications, including energy use, pollution, carbon footprint, land use, toxicity, and more. Many published life cycle assessments consider the entire cradle-to-grave approach by considering different use cases, material recycling, and recovery [421]. These assessments will be examined to compare the environmental and sustainability impacts of various electrode, electrolyte, and substrate materials being investigated for use in TSCs. Fig. 6 shows the general approach of LCA applied to assess a CNT manufacturing scheme for electronic applications.

Textiles can be spun and woven from manufactured or natural fibers derived from synthetic and natural resources, plants, and animals, respectively [9,422]. The more water, energy, land used, and emissions produced, the less sustainable a given fiber is. A material's biodegradability and recyclability also impact the sustainability of different fibers, because most textiles follow a linear life cycle from cradle to grave; they are usually discarded and sent to landfills after use by consumers or manufacturers [9,423]. In recent years, more importance has been placed on creating a circular life cycle of textiles by recycling them into new valuable materials to reduce or eliminate their contribution to solid

waste in landfills [424]. Experiments with extracting and repurposing the contents of wastewater produced by processing or dyeing textiles to eliminate this source of pollution or decrease cost of treatment are also being explored [425,426].

6.1. Environmental impacts of electrode and electrolyte materials

Energy storage devices generally use metals or carbonaceous materials in their electrodes; as a result, most of the energy consumption and environmental impact originate from materials preparation and processing. Comparing various TSC electrode materials, including graphene, conductive polymers, MXenes, and other metal oxides, reveals the superiority of these precursors, but production or end-of-life (EOL) strategies are needed. Unfortunately, EOL estimates are generally unavailable or widely assumed due to a data shortage for calculations and comprehensive estimates of toxicity impacts associated with release and waste handling in anticipated applications is known [427]. While nanoparticulate hazards are known for carbon nanotubes and graphene, their life cycle and toxicity are still being investigated [428,429].

From an emissions perspective, studies on graphene show that it not only improves the selected function of conductive textiles, but its use can also reduce environmental impacts. Cossuta reported that a graphene supercapacitor had a global warming potential (GWP) of 309 g carbon dioxide equivalents (CO₂Eq), over 80 % of which was from the production of the supercapacitor [421]. This value is insignificant when compared with the GWP of carbon composites like carbon fiber that have 54 kg CO₂Eq carbon footprint, significantly higher than other carbon-based electrode materials. The family of 2D transition metal carbides/nitrides, MXenes, were reported to have a significantly high energy demand for production, with titanium contributing more than 70 % of the adverse environmental effects due to greenhouse gas emissions [430,431]. This suggests that several adverse impacts could be addressed if recycled titanium was the primary metal source for Ti₃C₂T_x. Additionally, this energy consumption was defined based on synthesizing a 1 g batch of Ti₃C₂T_x MXene. The energy consumption for manufacturing kg- Ti₃C₂T_x MXene is expected to be one-ninth of the values reported for the production of MXene at 1 g batch [430–432].

Commonly used electrolytes in flexible TSCs can be integrated into LCA to compare the production and use cases. When investigating various electrolyte choices, it is hypothesized that aqueous electrolytes have LCA benefits compared to organic and ionic liquid systems [433]. Melzack's findings determined that the impacts of electrolytes cannot be reduced through substitution. Additionally, industrial plants often produce commonly used electrolytes that create toxic wastewater while synthesizing these materials. This wastewater can acidify local environments, adversely impacting the local ecology and human health.

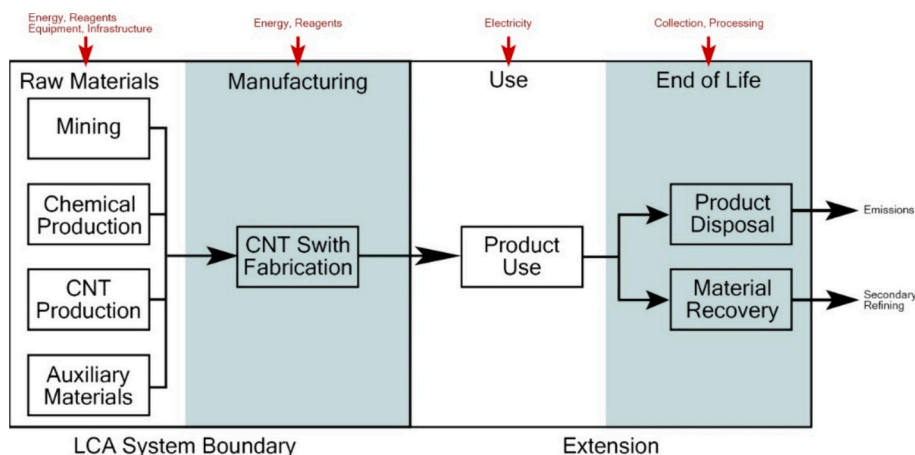


Fig. 6. Inputs and outputs of LCA for device fabrication using CNT. Adapted from reference [420].

These plants reduce this runoff by using circulatory water systems during electrolyte production [434,435]. Overall, recycling electrolytes at their end of life will play a role in reducing the overall impact [433].

Knowledge of LCA for various electrode materials for TSCs can help define strategies for mitigating their environmental impacts. For instance, using more renewable energy sources in materials preparation and processing and utilizing recycled metals could mitigate the environmental impact of producing metallic and carbonaceous components [430,431]. Researchers have also explored waste valorization for electrode materials, such as using pyrolysis to produce activated carbon from wastewater treatment sludge. This strategy could reduce the environmental impact of carbon-based materials used in flexible energy storage research [432].

Researchers have quantified the LCA of metal carbides and found that most of the energy used to synthesize these materials is used to produce their precursors. Most of the chemical impacts are caused by the metallic components in the metal carbides and the energy required for their production. Using recycled metals and renewable energy sources can mitigate the impacts of these materials in energy storage sectors [430].

6.2. Consideration of substrate materials

When investigating the LCA of TSCs, the 'Cradle-to-Grave' approach applies to the production, use, and EOL of the chosen substrate fibers. The most popular synthetic fibers are polyester and nylon, synthesized from petrochemicals derived from crude oil products and which comprise up to 64 % of all textile fibers produced in the world. Both fibers are synthesized by melt-spinning individual filament fibers into polymer chains before further processing into yarn and woven or knitted textiles; this process requires the least amount of energy and water and produces the least amount of carbon emissions, as shown in Table 6. Polyester and nylon require only the land used by industry to produce their filament fibers and textiles made from these derivatives; as a result, these fibers have a smaller land footprint than any natural fiber because there is no need to utilize land for cultivating their source material [422,426,436]. Toxic wastewater streams produced during the dyeing of these synthetic fibers are another source of pollution but can be mitigated or eliminated via dope dyeing the materials used to wet spin nylon and polyester fibers during the manufacturing of these textiles [437].

Nylon and polyester are easy to recycle as a part of efforts to create a circular textile economy and keep these materials out of landfills [422]. The recycling process becomes complicated for textiles when polyester or nylon is blended with another fiber; in order to recycle textiles, each fiber must be separated from all other fibers before they can be repurposed into new textiles [424]. Their recyclability also does not address or mitigate the microplastic fibers released into the aquatic

system by nylon and polyester textiles when they are washed [438]. These fibrous microplastics constitute up to 35 % of microplastic waste in the aquatic system, absorb toxic materials, and enter the marine life cycle; this toxicity can harm higher levels of the food chain due to the enrichment of prey with these toxic materials [438–440]. When considering the 'Cradle-to-grave' approach of LCA, the main environmental impacts in the use phase are caused by the washing, drying, and ironing of the garments. Washing 1 kg of fabric 50 times produces GHG emissions of 0.52 kg CO₂eq. Thankfully, green-conscious appliances and use habits can reduce energy use and CO₂ emissions. Reducing washing temperature from 60 to 40 °C saves approximately 40 % of electricity [441].

Natural fiber, such as silk, is an alternative fiber source for TSC. The *Bombyx mori* species of silkworm produces most of the world's commercially available silk. Silkworms hatch from an egg and feed on mulberry leaves until they mature, at which point each caterpillar spins itself a cocoon by wrapping itself in a single strand of silk fiber many kilometers long, making silk the only naturally occurring filament fiber [442]. The caterpillar is then either allowed to metamorphose into a moth to lay hundreds of eggs to replenish the population used to create silk textiles, meaning they are a renewable resource, or the cocoon is collected and boiled to kill the silkworm and start processing the cocoon into usable silk fiber. Silkworms that are killed are used as food, so nothing is wasted or left to decompose to produce methane, a greenhouse gas [9,442]. Most of the emissions, land, and energy required to produce silk textiles come from processing silk fiber; these emissions are offset by those absorbed by the mulberry trees that must be planted to feed the silkworms during their pupae and caterpillar life cycle stages, as shown in Table 6 [422,442,443]. Silk must first be degummed before it can be spun and dyed; both processes produce wastewater that can contaminate the environment, especially in the case of textile dyes [9,425]. Sericin recovery for pharmaceutical and cosmetic applications is also being explored to reduce biowaste from silk fiber processing and clean wastewater from the degumming process for reuse [426,444].

Silk textiles and garments face a similar cradle-to-grave problem that many other textiles do in that they are discarded after use instead of being reused, recycled, or repurposed [424]. To address this problem, regenerated silk fibroin (RSF) derived from discarded silk fiber during initial processing was created to reduce and eliminate this source of waste from the silk manufacturing industry and show that functional fibers could be made from this waste material source [9,445]. Remnants from processed silk textiles that had been discarded by garment manufacturers were also successfully used to create RSF fibers to reduce this source of waste by the silk industry [446].

Similar to silk, wool is produced by shearing the fur from sheep and processing the fiber into usable textiles. Processing wool also produces wastewater from cleaning and dyeing the fibers that can contaminate the environment. While wool comes from a renewable resource, the nature of concentrated animal farming requires more land to produce wool than what is required to cultivate other fibers, as shown in Table 6. Wool is also biodegradable and recyclable, but it is often mixed with other fibers such as cotton and nylon, which increases the difficulty of recycling wool garments and textiles because recycling textiles requires mixed fibers to be separated into their individual components before processing them into other textiles, as mentioned earlier [9,422,424,447]. Wool textiles also suffer from a linear cradle-to-grave life cycle, with efforts exploring how wool textiles can be repurposed or more efficiently recycled to create a circular economy and prevent this waste from going to landfill. Discarded wool textiles have been used as a source of fertilizer due to their biodegradability and the fact that wool is rich in nitrogen and sulfur [424,425,448]. Used and discarded wool garments have been utilized to produce new wool textiles for almost 200 years, creating a circular wool economy and decreasing the amount of wool disposal [449].

Cellulose occurs naturally in plants, so all cellulosic fibers utilize renewable resources and are biodegradable. Cellulosic fibers can be

Table 6

Comparison of energy, water, carbon emissions, and land use to produce various fibers.

Fiber	Energy (MJ/kg)	Water usage (kg/kg)	CO ₂ emissions (kg/kg)	Land use (ha/ton)	Ref.
Nylon	120	185	6.5	–	[422]
Polyester	125	62	2.8	–	[422]
Silk	1843 ^a	47,680 ^b	–63 ^c	14	[459–461]
Wool	63	165	2.2	278	[422,447]
Cotton	6	22,000	6	1.1	[422,447]
Viscose	100	640	9	0.7	[422,447]

^a Total energy required for mulberry tree growth, silkworm cocoon processing, and fiber processing.

^b Total water needed to grow mulberry trees, process silkworm cocoons, and process silk fibers.

^c Carbon emissions offset by mulberry tree growth.

collected and processed directly, as with cotton, the plants of which produce a cotton boll of cellulose fibers that can be harvested to produce cotton textiles [9,422]. These fibers can also be synthesized by wet spinning the cellulose extracted from wood pulp to produce polymer filament fibers such as viscose, with bamboo being a popular resource for this manufacturing process due to its rapid rate of growth [450–452]. Cellulose fibers are more difficult to dye than protein-based fibers, and cotton requires large amounts of water (as shown in Table 6) to raise cotton crop and process cotton fiber, and both activities produce a large amount of toxic wastewater [453]. While there are efforts to create more efficient and cost-effective methods of treating this effluent, others are exploring the elimination of this source of waste entirely. Cotton plants have been genetically modified to naturally grow pigmented cotton bolls to lessen or eliminate the need for dyed cotton textiles. This color-grown cotton will decrease the amount of pollution produced by dyeing cotton textiles and streamline the process of manufacturing these products. Streamlining the manufacturing process and introducing more cost-effective ways to manage wastewater in the cotton industry will help decrease the cost of manufacturing cotton textiles [454–456].

In addition to the wastewater produced to prepare cotton textiles, these products are typically discarded after use as garments, like many other textiles. To eliminate this contribution to landfills and create a circular economy, this discarded product has been sorted by color, dissolved into prepared dope, and wet spun into new fibers that closely resemble viscose. This eliminates discarded cotton clothing as a source of solid waste in landfills and eliminates the need to dye the new textiles made from this waste, as the dope is already pigmented [191,449]. Incorporating dye into wet spinning dope that utilizes freshly extracted cellulose has also been a method to produce pigmented viscose fibers that do not need to be dyed, eliminating the wastewater that would have been produced by the dyeing process [52]. This method streamlines the process of manufacturing viscose textiles by removing the time and labor that would have been necessary to dye viscose textiles, in addition to eliminating the cost of wastewater treatment. Both factors reduce the overall cost of production of viscose textiles [455].

Efforts can also be made to explore innovative approaches to design and manufacture new substrate materials, such as those made from biodegradable polymers and recycled textiles. Biodegradable polymers, like polylactic acid (PLA) and polyhydroxyalkanoates (PHA), offer the advantage of being derived from renewable resources and having minimal environmental impact [457]. PLA conversion releases about 2.9 kg of CO₂ per kg of PLA. An effective strategy for making PLA a low-carbon material can be optimizing its conversion process, as it consists of more than 50 % of the PLA greenhouse emissions in the landfilling and composting stages [458]. Incorporating recycled textiles from post-consumer waste or textile waste streams as substrates can also reduce the environmental burden of evaluating the life cycle impacts of different substrate materials, considering their production, usage, and end-of-life stages, will provide insights into their sustainability performance and guide the selection of materials with the lowest ecological footprints [422].

7. Biocompatibility of supercapacitors

The demand for biocompatible electrode and electrolyte materials as power sources for implanted medical devices has increased due to the increasing prevalence of these electronic systems. As a result, consideration of the biocompatibility of flexible supercapacitors has received more emphasis. TSCs have potential health care applications in chronic disease monitoring, remote therapies, electro-organ transplantations, and clinical disease state evaluations [462–466].

Wearable supercapacitors in implantable devices must meet requirements such as safety, prolonged life span, compact size, flexibility, stability without the need for packaging, lightweight, high reliability, high biocompatibility, and appropriate electrochemical performance [463,467]. Commercial supercapacitors cannot meet all of these

requirements and are currently prohibited from being integrated into implantable electronic devices due to their unstable and hazardous electrolytes, such as strong acids, bases, and flammable organics, as well as their weight and rigid configurations [468,469].

Researchers face many challenges that limit the correct functioning of implantable devices, such as difficulty in miniaturization, encapsulation, frequent replacement, inflammatory side effects in the body, and immunological rejection [51,470]. These problems can sometimes be significant enough to threaten the wearer's life. To this end, researchers around the world are working to develop and fabricate thin-film-based flexible and biocompatible supercapacitors with exceptional electrochemical performances, stable cyclic life spans, and no negative impact on the user.

7.1. Biocompatible electrolytes

The electrolytes used in wearable and flexible TSCs are usually toxic to some degree, but one way to mitigate this toxicity is to incorporate them into a sufficiently high-viscosity gel electrolyte and apply it to a TSC on the right side of fabric or clothing, preventing contact with the user [326]. Plasticizers are often incorporated into the gel electrolyte for safer incorporation into clothing to prevent the electrolyte from wearing away during washing, thus increasing the overall lifetime of the energy-storing garment [354]. Table 7 compares the performance of various flexible supercapacitors with the electrolytes used to assess their toxicity or danger to humans. The data in Table 7 shows that the researchers focus on studying the biocompatibility and toxicity of the textile-based supercapacitors.

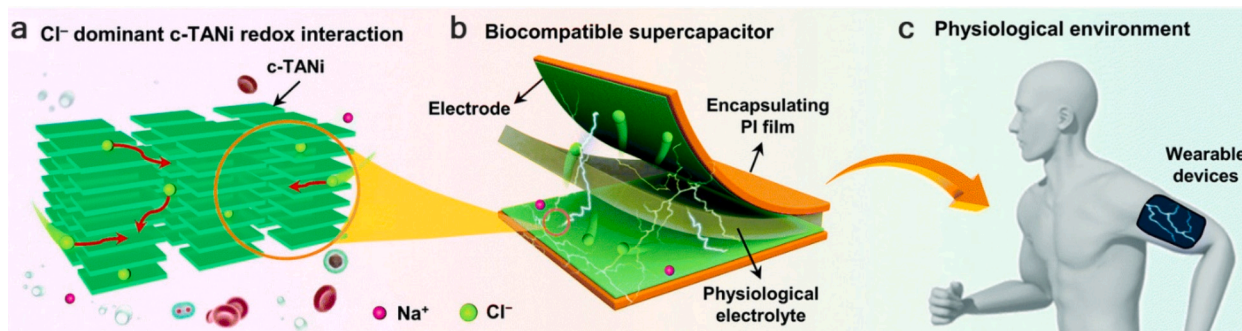
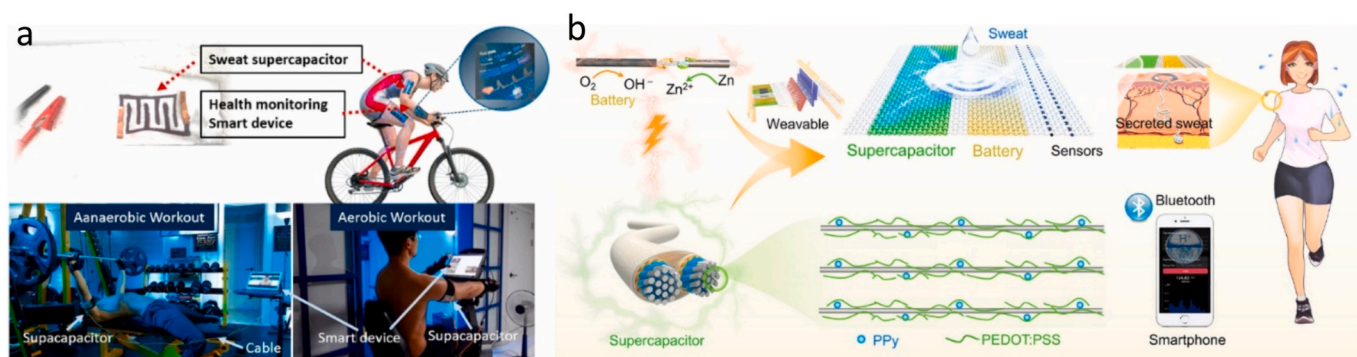
Several researchers focus on the use of physiological electrolytes in supercapacitors for wearable, epidermic, or even implantable applications (demonstrated in Fig. 5) to maintain living functions such as phosphate-buffered saline (PBS), serum, normal saline, sweat, blood, blood plasma, urine, and chloride-containing compounds (NaCl, KCl) [471,472]. For instance, Luo et al. [472] fabricated biocompatible CNT fibers as effective electrodes for supercapacitors with physiological fluids, including PBS, serum, and blood directly acting as electrolytes. A specific capacitance of 10.4 F/cm³ or 20.8 F/g was achieved for this device in PBS that could be maintained by 98.3 % after 10,000 cycles. These biocompatible fiber-shaped supercapacitors were lightweight, flexible, and miniature, and they could be further woven into soft electronic textiles aiming at a large-scale application. This work also provides a general and promising strategy for developing new materials and devices.

Xiaoling Tong et al. [473] reported a biocompatible system, shown in Fig. 7, including crystalline tetra-aniline (c-TANI) as the active electrode material and sodium chloride (NaCl) as a neutral physiological electrolyte. This device exhibited a remarkable areal capacitance (414 mF/cm² at a sweep rate of 5 mV/s) with 94.5 % capacitance retention even after 4000 cycles in a standard saline solution (0.9 wt% NaCl). This device also showed an areal energy density of 80.3 μW h/cm² with a power density of 0.82 mW/cm². A. Lamberti and C. F. Pirri [474] fabricated an implantable supercapacitor based on TiO₂ nanotube arrays as a biocompatible electrode and a PBS solution as a physiological electrolyte. This device demonstrated a specific capacitance of up to 1.42 mF/cm² at 0.01 V/s, an energy density of 0.18 μWh/cm², and a power density of 136 μW/cm². Recently, biofluids excreted from the human body, specifically sweat, have been utilized as natural electrolytes to prepare biocompatible energy sources due to their high ion contents and eliminating the need to encapsulate and isolate flexible energy storage systems from the user [475]. Sweat also has several advantages for biocompatible supercapacitors, such as non-toxicity, ecological safety, and reliability through electrolyte availability for wearable energy storage devices. The Libu Manjakkal research group [146] reported a sweat-based flexible supercapacitor including human sweat as an electrolyte and poly(3,4-ethylenedioxythiophene): poly(styrenesulfonate) (PEDOT: PSS) coated onto cellulose/polyester cloth

Table 7

The biocompatibility and toxicity performance of flexible supercapacitors.

Supercapacitor	Electrolyte	Capacitance	Toxicity	Biocompatible	Ref.
PEDOT: PSS coated cloth	Human sweat	3.88 F/g	None	Yes	[146]
Knitted 2D MXene-coated yarn	PVA/H3PO4	760 mF/cm ² ^a	Non-toxic on skin	–	[7]
PANI/CNT	PVA/H2SO4	1.6 mF/mm ²	No	–	[354,355]
MnO ₂ /CNT yarn	PVA/LiCl	61.25 mF/cm ² ^a	Yes	No	[99,143,356,357]
Graphene/CNT	PVA/H2SO4	177 mF/cm ²	No ^{b,c}	–	[69,355]
TiO ₂ /rGO	H2SO4	524 F/g	No	–	[175]
MWCNT/rGO	PMMA/ionic liquid ^d	30 mF/cm	No	–	[72]

^a Based on electrode cross-sectional area.^b Higher doses of graphene resulted in chronic toxicity.^c Refined CNTs did not result in a toxic response; their biocompatibility is thus far undetermined.^d Propylene carbonate (PC) 1-butyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide ([BMIM][TFSI]) ionic liquid.**Fig. 7.** Examples of biocompatible supercapacitor systems: a) c-TANI redox system, reproduced with permission [473], b) physiological electrolyte between two electrodes and an encapsulating film, c) applications for biocompatible TSCs in wearable devices.**Fig. 8.** a) Schematic demonstration of sweat textile-supercapacitor devices in action. Reproduced with permission [320]. b) Examples of flexible supercapacitors made with biocompatible electrolyte. Reproduced with permission [478].

as the active electrode. The performance of this device in artificial sweat exhibits a specific capacitance of 5.65 F/g, energy density of 1.36 Wh/kg, and power density of 329.7 W/kg. When tested under practical application, the sweat-based flexible supercapacitor was not as good as the performance under laboratory conditions. The performance of this device in real human sweat also decreases as evidenced by an energy density of 0.25 Wh/kg and a power density of 30.62 W/kg.

Selvam and Yim [320] fabricated a biocompatible textile supercapacitor based on PEDOT: TREN: MnO₂@MnCO₃ chelate decorated cotton fabric as an electrode and sweat as an electrolyte, see Fig. 8a. This device exhibits an impressive areal capacitance of 30 F/cm² and good capacity retention of about 92 % after 50,000 charge-discharge cycles. The biocompatibility tests of this material revealed that this sweat-based textile supercapacitor could be an excellent candidate to start fabricating textile supercapacitors for health monitoring in implantable devices. Ji and others [476] developed two biocompatible electrodes in supercapacitors by using the MnO₂/MWCNTs composite as the positive

electrode, phosphorated activated carbon as the negative electrode, and using the various natural ions (Na⁺, K⁺, Ca²⁺, Cl⁻, and HCO₃⁻) [477] of sweat as the electrolyte. This strategy takes advantage of body fluid directly as the electrolyte to avoid the need for encapsulation. Xiao et al. [478] developed a sweat-based yarn biosupercapacitor (SYBSC) with two symmetrically aligned electrodes made by wrapping hydrophilic cotton fibers on polypyrrole/poly (3,4-ethylenedioxythiophene): poly (styrenesulfonate)-modified stainless steel yarns. This artificial sweat system provides a high areal capacitance of 343.1 mF/cm² at 0.5 mA/cm² and a good capacity retention of about 68 % after 10,000 cycles. Zhiling Luo et al. [472] described the fabrication of sweat-chargeable on-skin supercapacitors by using a biocompatible NaCl electrolyte and free-standing polyaniline/carbon nanotube (PANI/CNT) electrodes. The device shows superior wearability because it can be directly attached to human skin and start the self-charging process by absorbing user sweat. Gschwind et al. [479] reported that a chloride-dominant neutral physiological electrolyte provides a solution to electrolyte safety concerns

while retaining adequate electrochemical performance.

7.2. Biocompatible electrode materials

Of equal importance is the biocompatibility of the electrode materials for wearable supercapacitors. Biocompatible material development is one of the most popular and promising avenues where TSC applications have been explored, with Fig. 8b showing potential applications of biocompatible electrolytes and electrodes. Conductive polymers such as PEDOT:PSS and Ppy are biocompatible, and utilizing sweat as an electrolyte is safer than many other electrolytes. To this end, several researchers have developed biocompatible materials to form supercapacitors as power sources for implantable electronics used for medical applications. Siddharth Sharma and others [470] fabricated a symmetric supercapacitor containing nanocluster di-titanium nitride ($n\text{-Ti}_2\text{N}$) thin films coated on a stainless steel 304 (SS-304) substrate as the electrode and PBS as a physiological electrolyte. This device showed outstanding electrochemical properties with an unprecedented areal capacitance of 11.62 mF/cm^2 , high areal energy density of $1.614\text{ }\mu\text{Wh/cm}^2$ with optimum areal power density of 49.98 mW/cm^2 , and a good capacitance retention of 86 % even after 5000 CV cycles at a scan rate of 50 mV/s . The biocompatibility of the electrode was checked in vitro, indicating excellent cell viability of around 85 %. These astounding outcomes indicated that this system makes a potential candidate for powering medical and implantable electronic devices by directly utilizing physiological fluid.

Sharma et al. [480] fabricated a flexible and biocompatible Niobium nitride ($\text{NbN}@SS||\text{TiN}@SS$ asymmetric supercapacitor in PBS. This system exhibits areal capacitance of 9.3 mF/cm^2 with excellent capacitive retention of 87.11 % after 10,000 cycles, superior energy, and power densities of $1.86\text{ }\mu\text{Wh/cm}^2$ and 239.14 mW/cm^2 , respectively, at 0.2 mA/cm^2 . The biocompatibility of the electrodes was checked in vitro, demonstrating exceptional cell viability of around 94 %. These remarkable results indicate that the fabricated device can be used in flexible and biocompatible supercapacitors in implantable medical applications.

Liu et al. [481] fabricated a stretchable hydrogel supercapacitor based on $\text{PANI}@r\text{GO}/\text{MXene}$ electrode and a hydrogel electrolyte. The system shows a high areal capacitance (45.62 F/g) and energy density ($333\text{ }\mu\text{Wh/cm}^2$, 4.68 Wh/kg). The in vitro and in vivo biocompatibility of the all-hydrogel micro-supercapacitor is evaluated by cardiomyocytes and mice models showing high biocompatibility. Mengqi Zhou and others [482] fabricated a biocompatible supercapacitor based on TiO_2 nanotubes as the electrode and PBS solution as the electrolyte. In this work, a biocompatibility assay that tracks cell proliferation on TiO_2 nanotube electrodes was discussed, and the device's biocompatibility was highlighted. The data revealed that the specific capacitance was $1040\text{ }\mu\text{F/cm}^2$. Biocompatibility tests and electrochemical characterization in a physiological liquid (0.01 M PBS solution) indicate that this system can be used as a power source for implantable and wearable devices. These results pave the way for biocompatible bioelectronic implants and could possibly benefit the development of bio-integrative electronics.

Furthermore, several researchers have recently used hybrid materials made of metal or carbon coated with non-toxic polymers to produce biocompatible electrodes for implantable supercapacitor devices. Sohini Chakraborty and others [483] prepared a biocompatible supercapacitor electrode based on styrene maleic anhydride (SMA) copolymer modified with a thiadiazole moiety along with the incorporation of ZnO nanoparticles. This device exhibits a specific capacitance of 268.5 F/g at 0.1 A/g and shows excellent cycling stability with maximum capacitance retention. The biocompatibility of this polymer nanocomposite electrode was studied by cytotoxicity tests. This study revealed that high biocompatibility can be used as an energy-storage device inside the human body, such as medical implants.

Selvam and Yim [319] described the fabrication of biocompatible

textile supercapacitor designed from chitosan/ $\text{MnO}_2@\text{MnCO}_3$ chelate-decorated fabrics. This device exhibits a remarkable areal capacitance of 2.5 F/cm^2 , an energy density of 0.35 Wh/cm^2 , and a good capacity retention of about 90 % after 10,000 charge-discharge cycles with good mechanical flexibility. Moreover, the biocompatibility of this material composite electrode was tested, and it revealed excellent cell viability of around 90 %. This result showed that these materials of textile supercapacitors can be used for health monitoring and treatment or wearable smart devices.

Hsu et al. [484] fabricated a mussel-inspired biocompatible hydrogel supercapacitor (TA-GelMA-CNC). The specific capacitance, energy density, and power density of the SCs reached 1861.21 mF/cm^3 , 20.65 mW/cm^3 , and 595.59 mWh/cm^3 , and retained 96 %, 100 %, and 82 %, respectively, of their original values after one cut-healing process. This system showed great potential for incorporation into energy storage devices for medical care. Chakraborty et al. [485] reported the fabrication of a wearable supercapacitor constructed from a copolymer nanocomposite containing ortho phenylenediamine and aniline incorporated with CNTs coated on fabric. This supercapacitor exhibited a high specific capacitance of 147.14 F/g at 0.50 A/g with a capacitance retention of 82 %. The biocompatibility of this nanocomposite polymer was examined by cytotoxicity tests and exhibited excellent biocompatibility. This study indicated great promise as a bio-supercapacitor in implantable medical devices.

8. Current challenges facing textile-based supercapacitors (TSCs), potential solutions, and future perspectives

Although TSCs have drawn significant attention, there are still several challenges that need to be addressed to ensure their reliability and user confidence. The electrical performance of TSCs must be relatively unaffected by repeated washing, and the materials used must withstand sustained and repeated mechanical deformation such as twisting, bending, and stretching. The effects of these mechanical deformations on the performance of TSCs must also be understood and quantified in order to design and fabricate wearable energy storage devices whose performance is either unaffected or improved by these physical distortions. Thus, novel approaches and methods for assessing composite electrodes' mechanical properties and their correlations with the state of charge and performance of TSCs are critical [486].

These challenges can be broadly categorized into material-related issues, manufacturing challenges, performance limitations, durability concerns, and sustainability and biocompatibility. Here, we present the main bottlenecks, potential solutions, and future perspectives.

8.1. Material-related issues

It is challenging to select suitable conductive materials and electrolytes that can be easily integrated into textiles while maintaining flexibility and conductivity [346,487]. Commonly used materials like carbon-based materials (graphene, carbon nanotubes) or conductive polymers often face issues like poor adhesion to textile fibers and degradation over time. The solution to this challenge is the development of advanced materials like flexible metal-organic frameworks, hybrid materials by nanocomposites, and conductive polymers with enhanced binding properties [52,488]. These hybrid materials can be incorporated into the textile fabric structure, maintaining flexibility and permeability and improving electrochemical performance over pure conductive polymers [19,487]. Electrolytes also need to be compatible with textile materials, providing good ionic conductivity without damaging the fabric. Solid or gel electrolytes are often preferred over liquid ones, but achieving high performance is difficult. In recent years, researchers have attempted to solve this challenge by developing solid-state or gel-based electrolytes compatible with textiles and having high ionic conductivity [346,363]. Incorporating ionic liquids in the TSC electrolytes can solve the challenges related to ionic conductivity and lead to improved

electrochemical performance [142,346,383].

Research into new materials that combine high conductivity with flexibility and durability will be crucial. Future research should focus on bio-compatible, sustainable, and environmentally friendly electrolytes that can be easily integrated into textiles without compromising their safety.

8.2. Manufacturing challenges and market standards for textiles

To achieve commercial viability, TSCs must also meet the market standards established by the American Society for Testing and Materials (ASTM) for everyday use [489]. Integration and scalability technologies are two of the most significant bottlenecks facing wearable textile supercapacitors. Techniques for uniformly incorporating energy storage materials into textiles while maintaining fabric flexibility and breathability must be improved. Ensuring uniform distribution of materials while maintaining the fabric's intrinsic properties is difficult. The solution to this challenge is the development of advanced fabrication and encapsulation techniques such as dip-coating, spray-coating, layer-by-layer technique, or electrospinning to improve uniform material distribution, maintain air permeability and improve the electrochemical performance of TSCs [83,490–492]. In recent years, some researchers have tried solving this challenge by exploring weaving or knitting conductive fibers directly into the fabric [141,314,493].

Creating scalable material systems while maintaining quality and performance in the production of textile supercapacitors is also a significant hurdle. Current manufacturing methods are often not suitable for mass production, and most reports show only a small quantity of scalable material systems being produced. To overcome this challenge, researchers and industrialists should adopt roll-to-roll processing and other scalable manufacturing techniques. If the authors can discuss how their process is scalable for industrial use, it will help readers understand the real-world implications of their research [494]. In addition to scalability issues when producing TSCs, this technology still has high manufacturing costs and little standardization in the manufacturing methods to fabricate them. Standardization of processes and materials can also help in achieving consistent quality. The research and development driving the integration of electronic components with clothing increases the cost of manufacturing these items, making them unavailable to some vendors [19]. Low-cost integration of supercapacitors into clothing can help to control the price, and consequently, TSCs will be more affordable.

Research into the development of manufacturing processes such as encapsulation materials, layer-by-layer technology and electrospinning techniques, along with the development of multi-functional textiles, will help in streamlining production and ensuring quality. Collaboration between academia and industry will also be essential to develop scalable manufacturing solutions. Experimental (pilot) production lines can help transition from lab-scale to industrial-scale production.

8.3. Electrochemical performance limitations

One of the most important challenges currently facing wearable textile supercapacitors is their electrochemical performance, such as energy density, power density, and cycling stability. In fact, the current electrochemical performance of wearable textile supercapacitors often falls short of meeting practical application requirements. Textile supercapacitors generally have lower energy densities compared to traditional batteries. Improving the energy storage capacity without compromising the fabric's properties is a major challenge. To overcome this challenge, researchers have tried to develop hybrid materials and electrolytes compatible with textiles to improve the electrochemical performance. Other researchers have attempted to develop advanced fabrication and encapsulation techniques to create layered structures with optimized thickness that can also enhance energy storage [83,490–492]. While supercapacitors are known for high power density,

balancing this with the structural integrity and comfort of the textile is complex. The solution to this challenge is to improve electrode and electrolyte design and material properties to enhance power density without compromising flexibility [346,487]. Another solution to this challenge is utilizing 3D structures in TSCs to increase surface area [495,496]. Ensuring long-term stability and electrochemical performance over many charge-discharge cycles is critical in wearable textile supercapacitors. Many current materials and designs degrade quickly with repeated use. Textile supercapacitors are also sensitive to environmental factors such as temperature changes and moisture, which can further impact their electrochemical performance and reliability. To overcome this challenge, researchers have tried to use robust materials and protective coatings to prevent degradation. Implementing self-healing materials could also enhance longevity in wearable textile supercapacitors [497–500].

Our perspective is that research innovations in electrode architecture and material designs will significantly improve the electrochemical performance, such as energy density, power density, cycling stability, and overall reliability, of textile-based supercapacitors. Future research should focus on optimized electrolytes and the development of materials and coatings that can self-repair or maintain stability under mechanical stress. Improving energy storage capacity (electrochemical performance) without compromising fabric properties through hybrid systems combining supercapacitors and batteries can also provide a balanced solution [501–505].

8.4. Durability concerns (washability and wearability)

Washability and durability are major hurdles currently facing wearable textile supercapacitors because most devices are delicate and fragile and have limited washability [47]. The main problem is the ability of wearable textile supercapacitors to withstand wet environments, sweat, and unfavorable conditions, such as in the case of washing machines and tumble dryers. Maintaining performance (long-term stability) after such mechanical stresses or environmental factors is a challenge in real-world conditions. TSCs should be able to withstand standard washing procedures employed by using a household washing machine [19,404]. In earlier reports, researchers did not address the washability of their proposed systems and thus did not discuss the best method to prevent deterioration as a result of washing [75,123,133]. In recent years, researchers have attempted to solve this challenge by removing the device from the textile before washing or encapsulating the supercapacitor device. The encapsulation strategy is one of the most frequently reported methods that protects both the electroactive materials and electrolytes from delaminating and leaking by coating an additional protective layer on the outside. Several research groups use silicone rubber, resin, and other polymeric materials to encapsulate the system. Encapsulation is also an effective way to use washable materials when constructing the electrode directly [328,420,506,507]. Solving the deterioration from washing and developing TSCs whose performance is relatively unaffected by stretching and bending during wear are critical problems to address to make TSCs break into the consumer clothing market.

Another challenge for wearable textile supercapacitors is long-term cyclability [494]. Many researchers wonder what will happen to the device during the lifetime of the garment; in practice, long-term cycling with good capacitance/capacity retention should be demonstrated. The second challenge facing wearable textile supercapacitors is durability due to their lack of stability against distortion, stretching, twisting, or bending. To achieve a long lifetime and applicable performance for wearable textile supercapacitors as power sources for many applications, the solution to this challenge is to design flexible and stretchable materials that can endure mechanical stresses.

Research into durable, moisture-resistant materials and encapsulation techniques will be essential to enhance environmental stability and mechanical durability. Future research should focus on developing

future textiles with an emphasis on durability and flexibility. Advances in material science and textile engineering will be pivotal.

8.5. Sustainability and biocompatibility

A few final challenges for wearable TSCs are sustainability and biocompatibility. The biocompatibility and sustainability of wearable TSCs must be quantified to assess their environmental impact and potential for medical applications before developing wearable TSCs as a product. The American Institute of Chemical Engineers (AIChE) has sustainability indices help assess a company's sustainability performance using the seven sustainability indices. These indices– sustainability innovation, strategic commitment, environment performance, safety performance, product stewardship, social responsibility, and value-chain management– reveal how the community, shareholders, customers, and peers perceive a company's sustainability efforts. Sustainability measurements are normalized as ratios of a measure of impact per unit mass, power, or economic value produced. Sustainability metrics that are applicable to a process, plant, or manufacturing system are material or energy intensity, potential environmental impact, or potential chemical risk. These metrics measure the number of nonrenewable materials or energy consumed per unit mass of primary product and economic value produced; they also measure the number of pollutants and amount of toxic emissions released to create the primary product. The biocompatibility and sustainability of wearable TSCs are influenced by fiber types, electrode and electrolyte materials, and synthesis methods [508–512].

To overcome these challenges, very few researchers have truly assessed the biocompatibility of supercapacitor systems [319,320]. Biocompatibility tests must be performed to verify that these materials and electrolytes in supercapacitors are non-toxic to humans because these systems will touch human skin and will be used as power sources for implantable medical devices. Research into developing biocompatibility and sustainability materials for electrodes and electrolytes in wearable textile supercapacitors will be essential to advance their use as power sources for implantable medical devices [513–515]. Once these challenges are addressed, TSCs will pave the way to change the future of smart textiles in wearable electronics, military devices, and implantable medical devices.

9. Conclusion

Flexible and wearable textile supercapacitors have attracted significant attention in recent years as energy storage devices for incorporation into smart textiles for various applications in medicine, sports science, and the military. The past decade has seen a boom in the development of wearable TSCs. Still, the manufacturing processes and material selections must be improved and streamlined before these devices become widely available. The main objective of this review article was to explore the materials and methods used in the development of wearable TSCs and to discuss their sustainability and biocompatibility. The opportunities and the need for research were highlighted, and the current state of the arts and the challenges were discussed. Lastly, the challenges related to washability, wearability, lifespan, biocompatibility, flexibility, and solid electrolytes must be addressed. Little research has been conducted into these areas and how they affect TSC performance.

From our point of view, creating practical and wearable textile-based supercapacitors requires several significant research efforts. The overview of current materials and fabrication methods reviewed in this work can help researchers working in energy storage to understand the fundamental challenges of TSCs and their future opportunities. In the future, energy management for personal use will become more important, with wearable devices to harvest waste energy becoming the norm, and the challenging missions needing direct human intervention, such as emergency and rescue missions, will depend on TSCs.

CRedit authorship contribution statement

Alyssa Grube: Writing – review & editing, Writing – original draft, Methodology, Formal analysis, Conceptualization. **Mahmoud M. Shaban:** Writing – review & editing, Writing – original draft, Investigation. **Laurel Hilger:** Writing – review & editing, Writing – original draft. **Mostafa Dadashi Firouzjaei:** Writing – review & editing, Visualization, Resources. **Ahmad Arabi Shamsabadi:** Writing – review & editing. **Yasar Demirel:** Writing – review & editing. **Mark Elliott:** Writing – review & editing, Visualization, Supervision. **Siamak Nejati:** Writing – review & editing, Writing – original draft, Conceptualization. **Mona Bavarian:** Writing – review & editing, Supervision, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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