Taylor & Francis
Taylor & Francis Group

Advanced Composite Materials

ISSN: (Print) (Online) Journal homepage: https://www.tandfonline.com/loi/tacm20

The use of fibre reinforced polymer composites for construction of structural supercapacitors: a review

Jayani Anurangi, Madhubhashitha Herath, Dona T.L. Galhena & Jayantha Epaarachchi

To cite this article: Jayani Anurangi, Madhubhashitha Herath, Dona T.L. Galhena & Jayantha Epaarachchi (2023) The use of fibre reinforced polymer composites for construction of structural supercapacitors: a review, Advanced Composite Materials, 32:6, 942-986, DOI: 10.1080/09243046.2023.2180792

To link to this article: https://doi.org/10.1080/09243046.2023.2180792

Published online: 02 Mar 2023.

Advanced Composite Materials, 2023
Vol. 32, No. 6, 942–986, https://doi.org/10.1080/09243046.2023.2180792

Taylor & Francis
Taylor & Francis Group

The use of fibre reinforced polymer composites for construction of structural supercapacitors: a review

Jayani Anurangi [a,b,c], Madhubhashitha Herath [b,d], Dona T.L. Galhena [e] and Jayantha Epaarachchi [a,b]

[a]*School of Engineering, Faculty of Health Engineering and Sciences, University of Southern Queensland, Toowoomba, Queensland, Australia;* [b]*Centre for Future Materials, Institute for Advanced Engineering and Space Sciences, University of Southern Queensland, Toowoomba, Queensland, Australia;* [c]*Department of Biosystems Technology, Faculty of Technological Studies, Uva Wellassa University of Sri Lanka, Passara Road, Badulla, Sri Lanka;* [d]*Department of Engineering Technology, Faculty of Technological Studies, Uva Wellassa University of Sri Lanka, Passara Road, Badulla, Sri Lanka;* [e]*Churchill College, University of Cambridge, Cambridge, UK*

Received 31 October 2022; accepted 08 February 2023

Fibre reinforced polymer plays an important role in many fields, especially in aviation and civil industries where lightweight design is a crucial factor. Over the past two decades, there has been extensive research on the development of multifunctional fibre reinforced composite structures which can fulfil several secondary functions besides its structural role. As a result, structural energy storage composites have been developing rapidly which can sustain electrochemical energy storage as well as structural load-bearing. Among the many structural energy storage composites, structural supercapacitor composites (structural supercapacitors) have attracted the attention of many researchers. This article provides an up-to-date review on the development of structural supercapacitors, which can be integrated into structural fibre reinforced polymeric components. Specifically, an outline is given of the development of carbon fibre fabric based structural supercapacitors, with the focus on various surface activations for performance improvement. Moreover, the recent development in critical components of structural supercapacitors, such as solid electrolytes and separators, is also highlighted. The limitations and challenges for the development of structural supercapacitors are also incorporated. Lastly, the novel fabrication processes and designs for future development are critically discussed. This article will help engineering and scientific communities to gain concise knowledge of structural supercapacitors.

Keywords: structural supercapacitor; energy storage; carbon fibre reinforced composite; carbon fibre electrode; multifunctional composite; structural electrolyte

1. Introduction

A steady and reliable supply of energy is essential for a modern industrial economy. The largest proportion of the world's energy is currently supplied by fossil fuels and, as a result, fossil fuel consumption has increased considerably during the last three decades. The limited

Corresponding author: Jayani Anurangi Jayani.Anurangi@usq.edu.au

sources of fossil fuels have resulted in mankind facing numerous energy problems [1]. Thus, there is a continuous interest in investigating renewable energies such as solar, wind, and tide as alternative sources of energy. However, the inherent characteristics of intermittence and uneven regional distribution of renewable energies demand high-performance energy storage devices. These energy storage devices charge when a surplus of energy is available and deliver it on demand when the source does not produce enough power. Nowadays, batteries, supercapacitors, and fuel cells are recognised as the three main systems for electrochemical energy storage [2] and are used in the civil and defence industries [3], e-mobility sector [4], smart electronics devices [5], and smart grids [6–8].

Currently, numerous studies are being undertaken to develop structural energy storage devices which can sustain mechanical loading while storing electrical energy [9,10]. Generally, the mechanical load-carrying capability and robustness of traditional energy devices are minimal. As a result, significant parasitic weight and space account for traditional energy storage systems. One such example is the conventional battery system of electric and hybrid vehicles [11]. Thus, the development of lightweight structures equipped with energy storage facilities is more desirable for electric and hybrid vehicles to save fuel, energy, and space [12,13].

Structural energy storage can be achieved in two ways: physical integration of standard energy storage devices into the traditional structural constituents [14–16] or functionalisation of the fibre reinforced laminate constituents for storing energy [17,18], as illustrated in Figure 1.

In various studies, it was reported that the former configuration, which has embedded energy storage devices, has many drawbacks compared with the latter configuration [14]. Particularly, the embedded energy storage devices in composites can reduce the stiffness, failure stress, fatigue strength, and other properties of composite materials [14,19]. In addition, these embedded devices cause discontinuity of the composite structure [20], and therefore the exertion of periodic load causes relative slippage between the layers. Due to the above problems, instead of physical integration of energy storage devices, the functionalisation of fibre reinforced composites for storing energy has gained favour as a way of realising structural energy storage composites.

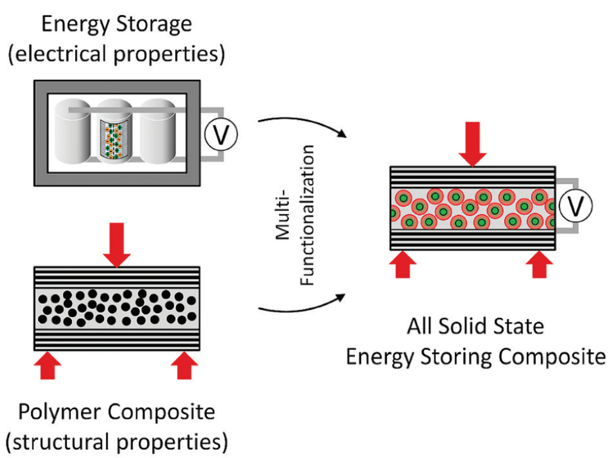

Figure 1. Concept of multifunctional energy storing composites with functionalised constituents [12].

Over the past decades, advanced composites such as carbon fibre reinforced polymers (CFRPs) have become popular in various structural applications because of their significant advantages such as a high specific strength-to-weight ratio and noncorrosive properties [21]. Nevertheless, carbon- based materials are suitable for electrochemical applications as they possess electrical conductivity [22,23].

Therefore, for advanced energy storage technologies, research interest in developing multifunctional composites with carbon fibre reinforcement has increased significantly [17,24,25]. Multifunctional energy storage composites which simultaneously carry a mechanical load while storing/delivering electrical energy enhance the system performance and efficiency by eliminating material redundancy. However, there is a trade-off between these two parameters since the improvement of one parameter will degrade the other parameter [26]. Thus, the development of a multifunctional composite is challenging due to this contradictory requirement of the constituents. As a result, optimisation of these two properties is essential when developing multifunctional energy storage composites [27,28].

With the rapid development in fibre reinforced laminates for storing energy, supercapacitor functional composite materials have attracted great attention as a leading candidate for an energy storage medium that has the ability to simultaneously store electrical energy and bear mechanical loads. In particular, the most common type of supercapacitors, the electric double layer capacitor (also known as EDLC), has the ability to maintain structural integrity and electrical properties due to its laminated structure and a charge-discharge mechanism which involves no physical destruction of electrode materials [29]. In addition, supercapacitor functional composites allow the incorporation of the energy storage function into any load-carrying part which requires electrochemical energy, for example, the body panels/chassis of electric or hybrid vehicles to power the engine, construction materials (wall bricks or roof tiles) to support the provision of electricity for households, and other potential applications.

The purpose of this review is to summarise the up-to-date development of structural supercapacitors (SSCs) which can be integrated into structural applications. As significant research has been carried out in this area, a timely review of such a rapidly growing field is highly desirable. Many researchers have focused on the development of three major components of supercapacitor functional composites, namely the carbon fibre-based electrode, the structural electrolyte, and the separator, and focused on various fabrication methods. These aspects have attracted our interest and led to this review article. Moreover, experimental results shown in the literature and common methodologies to optimise overall performance are presented in this article. Then, the existing technical challenges for the development of the main components of SSCs are summarised. Following this, novel fabrication processes and novel designs for future development are discussed in the latter part of the article. As a promising novel design, sandwich composite structure is described in detail. In addition, prospects for application are outlined. Furthermore, the sustainability of the outcomes to date is critically investigated and discussed in this review.

2. Recent developments in structural supercapacitors

The constituents of SSCs simultaneously and synergistically perform two roles: electrical energy storage and structural support, though it is challenging to balance these two conflicting requirements [30]. As a single engineering structure, these dual functional devices utilise materials efficiently and have the potential to achieve weight-to-volume gain in various structural applications [31]. For example, dual functional energy storage supercapacitors allow the conversion of the whole engineering structure into an energy storage device, resulting in the saving of space and mass.

Generally, an SSC consists of two robust electrodes separated by a thin film (separator), and a structural electrolyte that provides both mechanical and electrical properties. Many studies have been devoted to developing SSCs using various materials and architectural designs. SSCs based on carbon fibre reinforced material have gained a great deal of attention in the past [5,32–34], but room for overall improvement still remains. The common laminated architecture of the structural supercapacitor, fabricated with electrically conductive carbon fibre (CF) weaves as reinforced electrodes and glass fibre (GF) weave as the separator, is simply demonstrated in Figure 2.

SSCs are typically divided into three categories according to their energy storage mechanism: electrochemical double layer capacitors (EDLCs), pseudocapacitors, and hybrid capacitors. In an EDLC, energy is stored due to the accumulation of pure electrostatic charge in the electrode-electrolyte interface, as shown in Figure 3a. Activated carbon materials, carbon nanofibres, and graphene are commonly coated on CF fabric and used as electrodes of EDLCs. In pseudocapacitors, on the other hand, energy is stored due to electron transfer between the electrolyte and the electrode through reversible faradaic redox reactions, as illustrated in Figure 3b. Pseudocapacitors have a limited lifetime compared to EDLCs, due to chemical depletion at the surface of electrodes during the charging and discharging process. For pseudocapacitors, metal oxides such as MnO_2, NiO, TiO_2, polyaniline, and other conducting polymers are coated on the CF fabric. Unlike EDLCs and pseudocapacitors, the hybrid supercapacitors are made from two different types of electrodes. Therefore, hybrid supercapacitors (Figure 3c) combine the advantages of both pseudocapacitors and EDLCs. Their energy density is higher than EDLCs and their cyclic stability is higher than pseudocapacitors [35].

2.1 *Reinforced electrodes*

Among the various types of materials used, CF fabric is one of the most popular electrode materials for SSCs because it possesses both electrical conductivity and excellent mechanical properties [32,36,37]. CF eliminates the need for additional

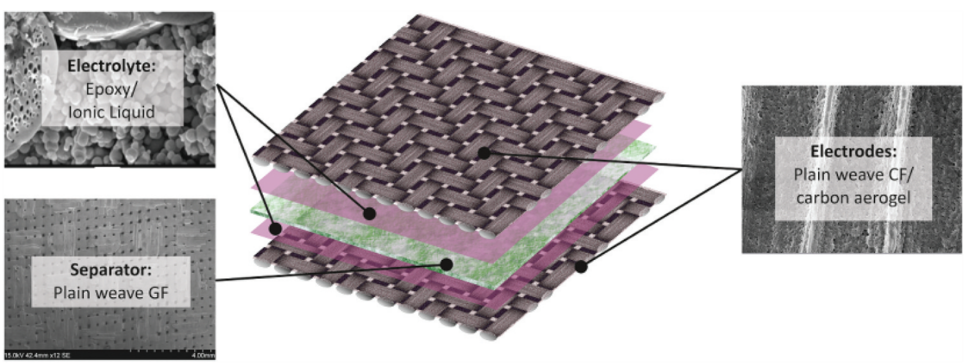

Figure 2. A schematic configuration of a structural EDLC [38].

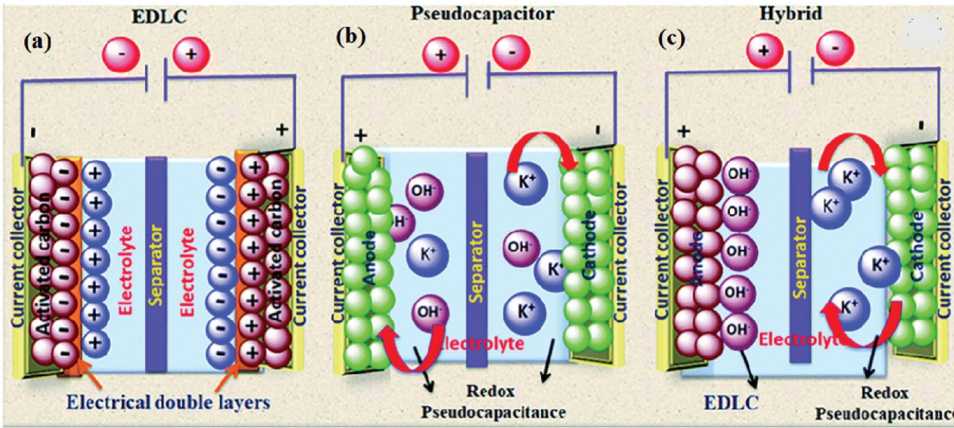

Figure 3. The charge storage mechanism of (a) EDLC, (b) pseudocapacitor, (c) hybrid capacitor [39].

substrates or current collectors as it possesses electrical conductivity. Further, CF is suitable as a reinforcement as it has high strength and stiffness. Recently, a great deal of effort has been invested in the development of electrodes using carbon fibre fabrics [5,32–34,40–45]. Typically, CF composites fabricated with pristine carbon fibre fabrics/weaves without modifications are unable to provide the expected electrochemical outcome, due to low surface area [22]. Modifications of CF increase the specific surface area by introducing mesopores at the surface of fibres. There are several methods used by researchers to modify the surface of CF fabric (Figure 4a). The first approach is physical and chemical surface activations, using steam, carbon dioxide, alkalis, and acids, to improve the electrochemical performance of CF fabrics [46]. Chemical activation is widely used to improve the electrochemical performance of carbon fibre fabric without significantly degrading the load-bearing capability. However, if the amounts of activation agents and process parameters (such as soak time) are not properly controlled, the burn-off level of carbon fibre might be exceeded, resulting in low mechanical properties [32].

Alternatively, CF surface covering with suitable electrochemically active materials with a large specific surface area is found to lead to a considerable improvement in electrochemical properties [47]. Carbon materials, such as activated carbon [48], carbon nanotube (CNT) [18,48], carbon aerogel (CAG) [33,49,50], graphene and its derivatives such as graphene nanoflakes [45], graphene aerogel (GA) [50], and graphene nanoplatelets (GNP) [24,52] can be coated on the surface of carbon fibre to enhance the EDLC behaviour. Several studies have shown that graphene-based materials are more suitable due to their high mechanical strength, the comparative ease of dispersion, high surface area, and high conductivity [51,53–55]. The other possible alternative is coating the conductive polymer or metal oxides to provide pseudocapacitance behaviour. Figure 4b summarises the improvement of the specific surface area of carbon fibres after various surface modifications.

When CF fabric is covered with electrochemically active materials, it might affect to the mechanical properties of its composite. Therefore, Artigas-Arnaudas et al. studied the effect of three types of GNP coated CF fabric on the mechanical properties of its CFRP

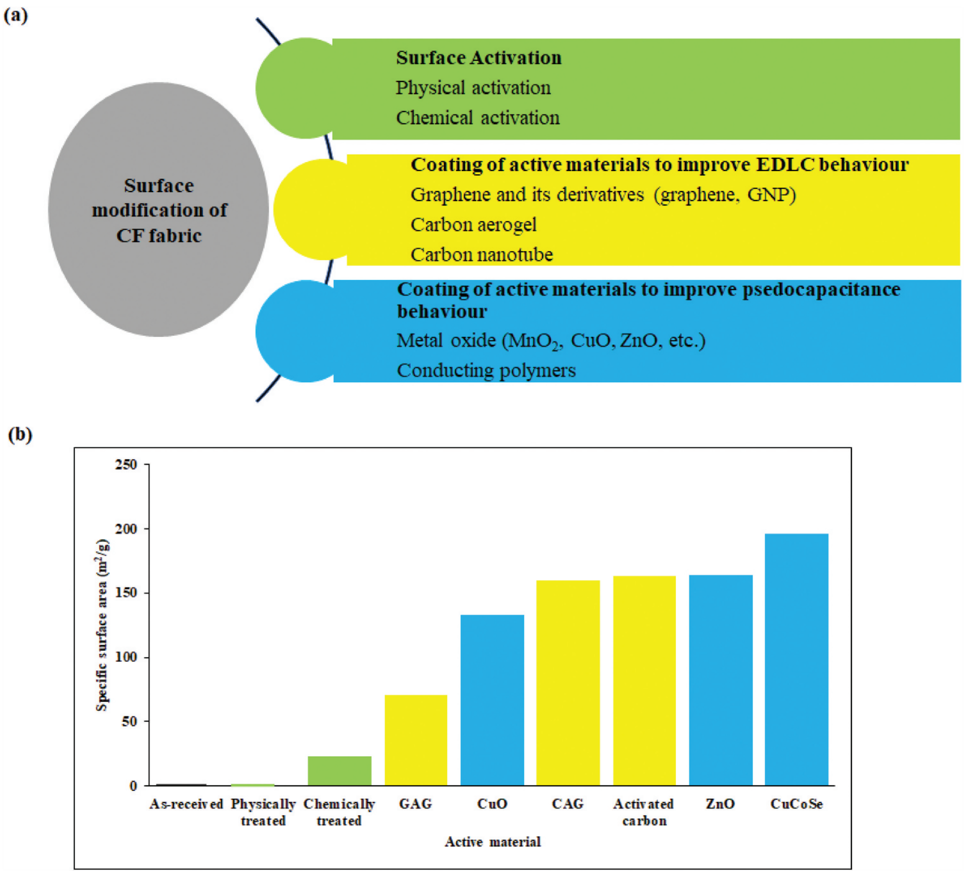

Figure 4.: (a) Modification methods for carbon fibre electrodes, (b) Surface areas of carbon fibres after modifications, data from reference lists of [3,5,32,40,52,56].

(see Figure 5). For this, they prepared CF electrodes with different GNP coating conditions (without any binder, with PVA binder, and with PVDF binder) and carried out interlaminar shear strength tests (ILSS) for the corresponding CFRP. Tests has been conducted accordingly to ASTM D2344. It has been shown that the deposition of GNPs is not severely detrimental to mechanical properties.

To improve the performance of CF electrodes, Deka et al. modified the carbon fibre fabric by growing CuO using a hydrothermal process. The specific surface area of the CuO grown CF was increased to 132.85 $m^2.g^{-1}$ (electrode mass specific) after this modification [40]. The specific capacitance of the fabricated supercapacitor with the CuO nanowire-grown woven CF electrodes, polyester resin (PES) matrix, and woven GF separator was as high as 2.48 $F.g^{-1}$. However, this value increased twofold (5.48 $F.g^{-1}$) after ionic liquid (1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl) imide (EMIMBF4)) was mixed into the PES matrix and it increased threefold (6.75 $F.g^{-1}$), when a lithium salt (Lithium trifluoromethanesulfonate (LiTf)) was added to the polyester ionic liquid mixture.

In addition, Javaid et al. investigated a structural supercapacitor with GNP incorporated CF fabric as the electrodes, diglycidyl ether of bisphenol-A (DGEBA) epoxy as the

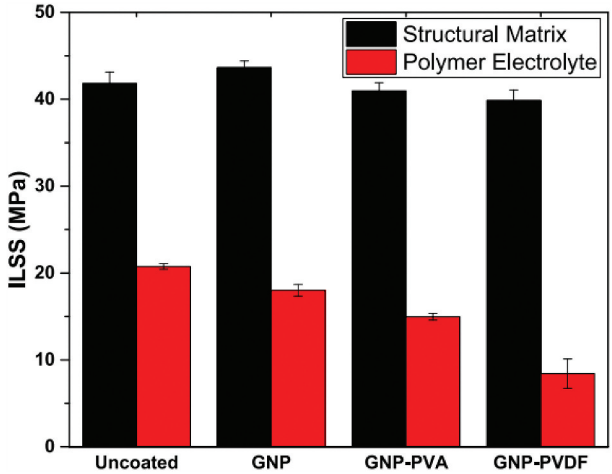

Figure 5. ILSS values for the different conditions tested, the black columns denote the structural resin and the red ones, the solid polymer electrolyte [57].

electrolyte, and filter paper (Grade 1, 0.1 mm layer thickness, Whatman) as the separator [52]. They fabricated the structural supercapacitor as illustrated in Figure 6. Both the mechanical and electrical properties of GNP-loaded carbon fibre-based SSCs showed obvious improvements by 13-fold compared with the control sample composed of pristine carbon fibre electrodes.

When comparing the performance of this device with the previous [42,58] and later work [3] done by Javaid and his group, high electrical properties with acceptable mechanical performance were reported for it [52]. Therefore, the loading of high surface area GNPs on carbon fibre fabric is an effective way to scale up for the improved performance of the electrodes.

In addition, Javaid et al. [41] studied performance improvement by investigating the graphene nanoplatelet-incorporated carbon aerogel coated CF-based structural supercapacitors. The SEM images, as shown in Figure 7, proved that the surface area of the

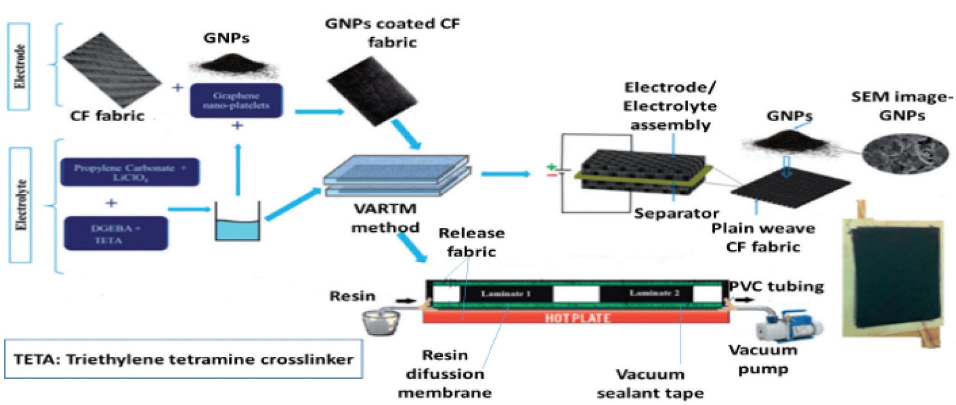

Figure 6. Schematic of fabrication process of GNP incorporated CF structural supercapacitor [52].

CF electrode significantly increases with the increase of GNP loading (1% to 5%). As a result, as illustrated in Table 1, its electrical performance was improved 10-fold after GNP coating (5% wt GNP).

Recently, Subhani et al. prepared graphene aerogel (GAG) impregnated carbon fibre fabric as the electrode material for SSC applications [51]. The supercapacitor was fabricated by using an electrospun nano-veil separator and ionic liquid modified epoxy-based electro-

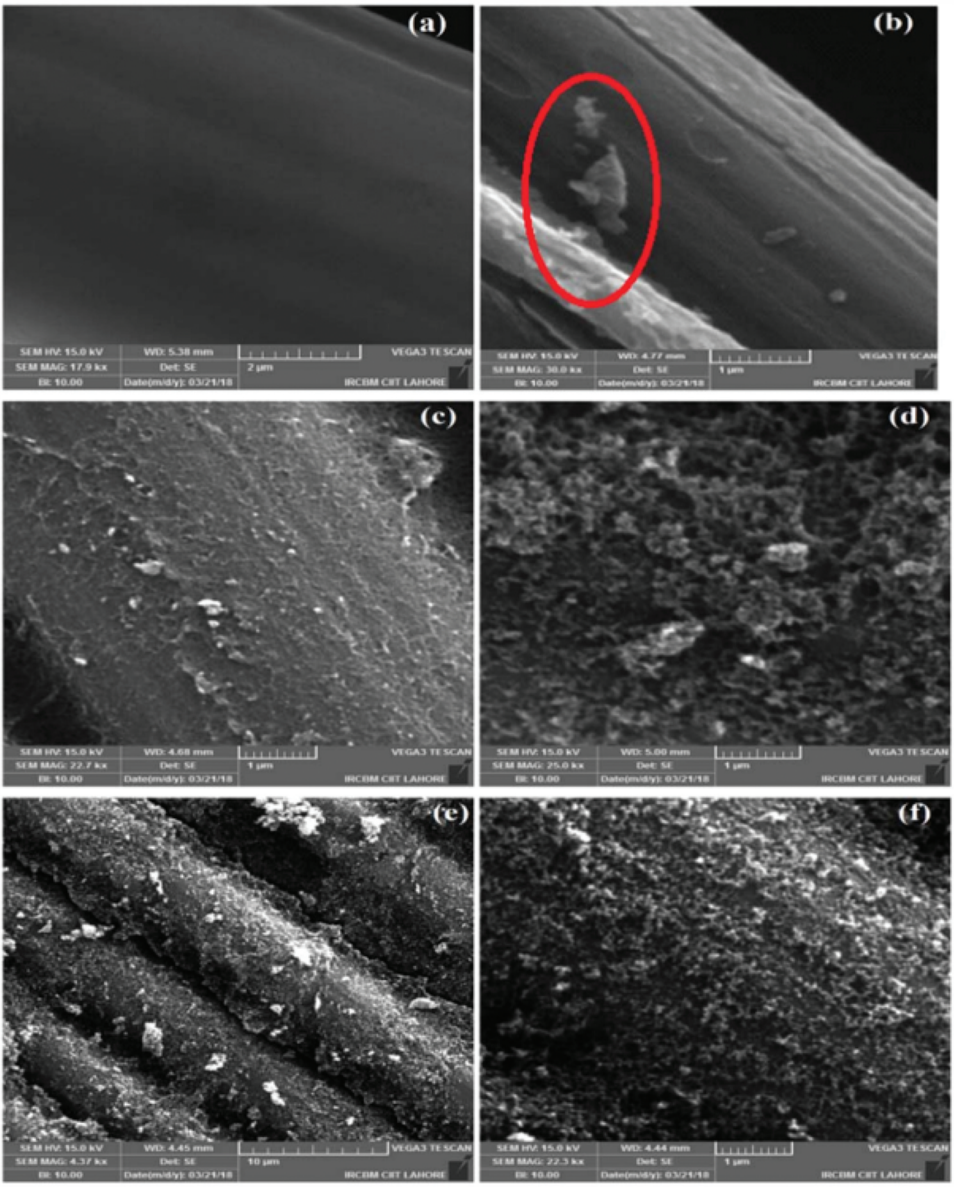

Figure 7. SEM images of (a) as received CF, (b) carbon aerogel coated CFs, (c) CF/CAG.GNP1, (d) CF/CAG.GNP3, (e) CF/CAG.GNP5 at 22000x, (f) 5000x magnification [41].

lyte. Subhani et al. demonstrated that the fabricated supercapacitor functional composite with 28% GAG loading on carbon fibre fabric had a capacitance of 56 $mF.g^{-1}$ (electrode mass specific) according to the CV test and a power density of 22.5 $mW.kg^{-1}$.

Besides, Javaid et al. investigated the electromechanical properties of a supercapacitor fabricated with polyaniline (PAni) deposited CF fabric electrodes, as shown in Figure 8. A filter paper was used as the separator and epoxy-based polymer electrolyte (DGEBA mixed with lithium salt (lithium perchlorate ($LiClO_4$) was used as the electrolyte. The resin infusion under flexible tooling method was used for the supercapacitor fabrication.

Here, as received and chemically activated CF fabrics after in-situ PAni deposition were investigated as electrodes of symmetric supercapacitors. Increasing the coating density of PAni on the CF fabric increased the multifunctionality of the supercapacitor. However, by increasing the coating density beyond 0.05 $mg.cm^{-2}$, there was a significant drop in both the specific capacitance and specific energy. The fabricated supercapacitor with PAni deposited activated carbon fibre electrodes exhibited 22.2 $mF.g^{-1}$ specific capacitance (device normalised) and 49.4 $mWh.kg^{-1}$ specific energy with 21.63 $W.g^{-1}$ specific power. In addition, the mechanical properties of the supercapacitor were measured as 1.1 GPa shear modulus and 6.3 MPa shear strength. Table 1 shows more detail about the electrical and mechanical performance of SSCs with modified CF electrodes, developed by different researchers.

In summary, every researcher has shown that modified CF electrodes have high electrical properties compared to pristine CF electrodes. In particular, graphene nanoplatelet-based CF electrodes showed an improvement in electrical properties without a severe detriment to mechanical properties. However, the resulting electrical and mechanical properties are still not adequate for use as electrodes of SSCs for structural applications. Therefore, it is clear that the investigation of further modification of carbon fibre fabric is needed to enhance its electrical properties while maintaining its load-bearing capabilities.

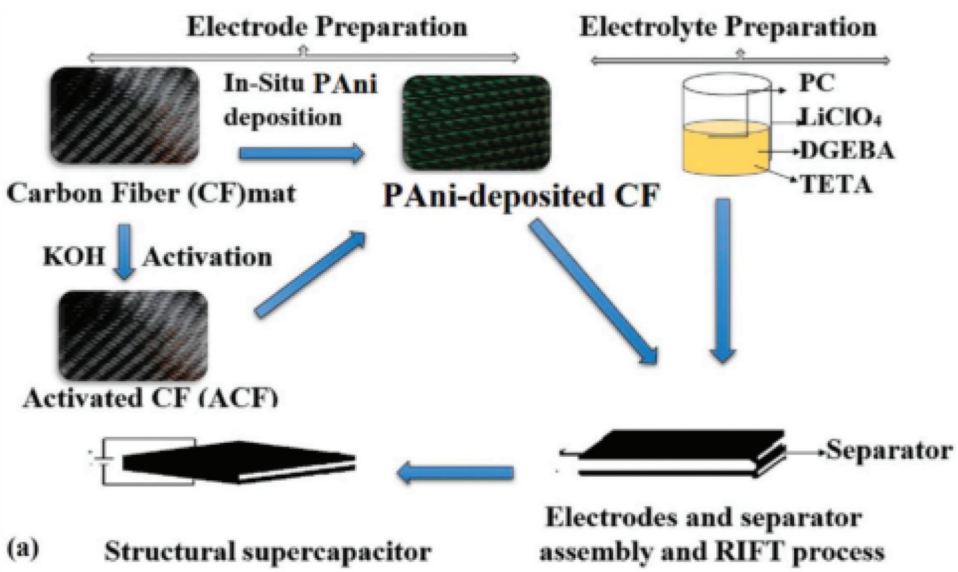

Figure 8. Schematic illustration of steps used for fabrication of the structural supercapacitor with PAni deposition [3].

Table 1. Electrical and mechanical properties of carbon fibre-based SSCs.

Item number		Configuration of structural supercapacitor			Electrical properties			Mechanical properties	Ref
		Electrode	Structural electrolyte	Separator	Electrode mass normalised	Device normalised	Mode of measurements		
01	i	CF	PEGDGE+IL (EMITFSI)	2GF	$C = 10.7$ $mF.g^{-1}$ $E_d = 0.015$ $mWh.kg^{-1}$ $P_d = 0.003$ $W.kg^{-1}$	Not reported	CA test, applying 0.1V for 60s	$\tau = 5.36$ MPa, $G = 0.3$ GPa	[33]
	ii	CAG-CF	PEGDGE+IL (EMITFSI	2GF	$C = 71.2$ $mF.g^{-1}$ $E_d = 0.10$ $mWh.kg^{-1}$ $P_d = 0.004$ $W.kg^{-1}$			$\tau = 8.71$ MPa, $G = 0.9$ GPa	
	iii	CF	PEGDGE+IL (EMITFSI)	PP	$C = 6.52$ $mF.g^{-1}$ $E_d = 0.009$ $mWh.kg^{-1}$ $P_d = 0.002$ $W.kg^{-1}$			Not reported	
	iv	CAG-CF	PEGDGE+IL (EMITFSI)	PP	$C = 602.5$ $mF.g^{-1}$ $E_d = 0.8$ $mWh.kg^{-1}$ $P_d = 0.03$ $W.kg^{-1}$			Not reported	

(continued)

Table 1. (*Continued*).

Item number		Configuration of structural supercapacitor				Electrical properties		Mechanical properties	Ref
02	i	CF	PEGDGE +LiTFSI	GF	Not reported	$C = 4.5\ mF.g^{-1}$ $E_d = 0.2\ mWh.kg^{-1}$ $P_d = 8.82\ W.kg^{-1}$	CA test, applying 0.1V for 60s	E= 21.70 GPa	[31]
	ii	ACF	PEGDGE +LiTFSI	GF		$C = 1.4\ mF.g^{-1}$ $E_d = 0.003\ mWh.kg^{-1}$ $P_d = 0.05\ W.kg^{-1}$		E= 32.03 GPa	
	iii	CF	PEGDGE +IL (EMITFSI)+ LiTFSI	GF		$C = 11.1\ mF.g^{-1}$ $E_d = 0.26\ mWh.kg^{-1}$ $P_d = 8.33\ W.kg^{-1}$		E= 15.71 GPa	
	iv	ACF	PEGDGE +IL (EMITFSI)+ LiTFSI	GF		$C = 52.2\ mF.g^{-1}$ $E_d = 1.43\ mWh.kg^{-1}$ $P_d = 2.68\ W.kg^{-1}$		E= 18.04 GPa	

(*continued*)

Table 1. (*Continued*).

Item number		Configuration of structural supercapacitor			Electrical properties			Mechanical properties	Ref
03	i	CF	PEGDGE+ LiTFSI	FP	Not reported	C = 0.21 $mF.cm^{-3}$	CA test, applying 0.1 V for 40s	τ = 7.3 MPa, G = 0.38 GPa	[42]
	ii	ACF	PEGDGE+ LiTFSI	FP		C = 0.41 $mF.cm^{-3}$		Not reported	
	iii	CF	PEGDGE+DGEBA+ LiTFSI	FP		C = 0.1 $mF.cm^{-3}$		τ = 8.51 MPa, G = 0.42 GPa	
04	i	CF	PEGDGE+IL (EMITFSI)	GF	Not reported	C = 10.3 $mF.cm^{-3}$	CA test, applying 0.1 V for 600s	τ = 6.12 MPa, G= 0.35 GPa	[58]
	ii	CF	PEGDGE+IL (EMITFSI)	PP		C = 11.9 $mF.cm^{-3}$		τ = 4.83 MPa, G= 0.31 GPa	
	iii	CF	PEGDGE+IL (EMITFSI)	FP		C = 7.1 $mF.cm^{-3}$		τ = 8.01 MPa, G= 0.42 GPa	
05		CuO-CF	PES+IL (EMIMBF4)+ LiTf	GF	C = 6750 $mF.g^{-1}$ E_d = 106.0 $mWh.kg^{-1}$ P_d = 12.6 $W.kg^{-1}$	Not reported	CV test, -0.5 V to 0.5 V, scan rate 10 $mV.s^{-1}$, three electrode system	σ = 251.8 MPa, E = 19.6 GPa τ = 92.6 MPa, G = 18.9 GPa	[40]

(*continued*)

Table 1. (*Continued*).

Item number		Configuration of structural supercapacitor			Electrical properties			Mechanical properties	Ref
06	i	CF	PES	GF	C = 213 $mF.g^{-1}$ E_d = 26.4 $mWh.kg^{-1}$ P_d = 1.0 $W.kg^{-1}$	Not reported	CV test, -0.5 V to 0.5 V, scan rate 10 $mV.s^{-1}$, two electrode system	σ = 154.9 MPa, E = 11.0 GPa τ = 63.3 MPa, G = 7.9 GPa	[56]
	ii	ZnO-CF	PES+IL (EMIMBF4)+ LiTf	GF	C = 7810 $mF.g^{-1}$ E_d = 81.2 $mWh.kg^{-1}$ P_d = 9.0 $W.kg^{-1}$			σ = 221.8 MPa, E = 15.9 GPa τ = 95.8 MPa, G = 12.0 GPa	
	iii	ZnO-CF	PES+IL (EMIMBF4)+ LiTf+ polyaniline nanofibre	GF	C = 18820 $mF.g^{-1}$ E_d = 156.2 $mWh.kg^{-1}$ P_d = 19.9 $W.kg^{-1}$			σ = 325.8 MPa, E = 21.6 GPa τ = 128.8 MPa, G = 15.6 GPa	

(*continued*)

Table 1. (*Continued*).

Item number		Configuration of structural supercapacitor			Electrical properties			Mechanical properties	Ref
07	i	CF	DGEBA+$LiClO_4$	FP	Not reported	C = 8.9 $mF.cm^{-3}$ E_d = 0.02 $mWh.cm^{-3}$	CA test, applying 0.1V for 400s	τ = 11.7 MPa, G = 1.4 GPa	[52]
	ii	GNP-CF	DGEBA+$LiClO_4$	FP		C = 118.7 $mF.cm^{-3}$ E_d = 0.26 $mWh.cm^{-3}$		τ = 19.1 MPa, G = 2.8 GPa	

(*continued*)

Table 1. (*Continued*).

Item number		Configuration of structural supercapacitor			Electrical properties			Mechanical properties	Ref
08	i	CF	DGEBA+$LiClO_4$	FP	Not reported	C = 35.7 $mF.g^{-1}$ E_d = 79.4 $mWh.kg^{-1}$ P_d = 13.1 $W.kg^{-1}$	CA test, applying 0.1V for 1500s	τ* = 3.6 MPa, G = 1.4 GPa	[41]
	ii	CAG-CF	DGEBA+$LiClO_4$	FP		C = 80.8 $mF.g^{-1}$ E_d = 179.6 $mWh.kg^{-1}$ P_d = 17.7 $W.kg^{-1}$		τ *= 3.8 MPa, G = 1.5 GPa	
	iii	CAG -GNP (1 wt%)-CF	DGEBA+$LiClO_4$	FP		C = 85.4 $mF.g^{-1}$ E_d = 189.8 $mWh.kg^{-1}$ P_d = 19.8 $W.kg^{-1}$		τ* = 4.8 MPa, G = 1.5 GPa	
	iv	CAG -GNP (3 wt%)-CF	DGEBA+$LiClO_4$	FP		C = 227.9 $mF.g^{-1}$ E_d = 506.5 $mWh.kg^{-1}$ P_d = 46.0 $W.kg^{-1}$		τ *= 5.2 MPa, G = 1.6 GPa	
	v	CAG -GNP (5 wt%)-CF	DGEBA+$LiClO_4$	FP		C = 353.7 $mF.g^{-1}$ E_d = 786.0 $mWh.kg^{-1}$ P_d = 107.8 $W.kg^{-1}$		τ* = 6.7 MPa, G = 2.0 GPa	

(*continued*)

Table 1. (*Continued*).

Item number		Configuration of structural supercapacitor			Electrical properties			Mechanical properties	Ref
09	i	CF	PEGDGE+IL (EMITFSI)	GF	Not reported	C = 12.7 $mF.cm^{-3}$ E_d = 12.2 $mWh.kg^{-1}$ P_d = 18.0 $W.kg^{-1}$	CA test, applying 0.5V for 1500s	τ = 6.1 MPa, G = 0.4 GPa	[59]
	ii	ACF	PEGDGE+IL (EMITFSI)	GF		C = 92.1 $mF.cm^{-3}$ E_d = 88.0 $mWh.kg^{-1}$ P_d = 14.2 $W.kg^{-1}$		τ = 6.2 MPa, G = 0.3 GPa	
	iii	CF	PEGDGE+IL (EMITFSI) +MSP	GF		C = 21.5 $mF.cm^{-3}$ E_d = 20.5 $mWh.kg^{-1}$ P_d = 35.2 $W.kg^{-1}$		τ = 39.4 MPa, G = 1.5 GPa	
	iv	ACF	PEGDGE+IL (EMITFSI) +MSP	GF		C = 120.4 $mF.cm^{-3}$ E_d = 117.7 $mWh.kg^{-1}$ P_d = 34.4 $W.kg^{-1}$		τ = 38.6 MPa, G = 1.8 GPa	

(*continued*)

Table 1. (*Continued*).

Item number		Configuration of structural supercapacitor			Electrical properties			Mechanical properties	Ref
10	i	CF	PES+IL (EMIMBF4)+ LiTf	KF	$C = 197$ mF.g^{-1} $E_d = 24.6$ mWh.kg^{-1} $P_d = 0.9$ W.kg^{-1}	Not reported	CV test, -0.5 V to 0.5 V, scan rate 10 mV.s^{-1}, two electrode system in 3M KCl	$\sigma = 258.2$ MPa, $E = 19.2$ GPa	[5]
	ii	Co selenide nanowires-CF	PES+IL (EMIMBF4)+ LiTf	KF	$C = 10920$ mF.g^{-1} $E_d = 105.6$ mWh.kg^{-1} $P_d = 11.24$ W.kg^{-1}			$\sigma = 434.1$ MPa, $E = 24.0$ GPa	
	iii	Cu selenide nanowires-CF	PES+IL (EMIMBF4)+ LiTf	KF	$C = 14760$ mF.g^{-1} $E_d = 136.4$ mWh.kg^{-1} $P_d = 16.9$ W.kg^{-1}			$\sigma = 443.9$ MPa, $E = 258$ GPa	
	iv	Cu-Co selenide nanowires – CF	PES+IL (EMIMBF4)+ LiTf	KF	$C = 28630$ mF.g^{-1} $E_d = 191.6$ mWh.kg^{-1} $P_d = 36.6$ W.kg^{-1}			$\sigma = 488.9$ MPa, $E = 32.6$ GPa	
11		ACF	Epoxy+ 1 M $TEABF_4$ in PC	CP	Not reported	$C = 25.4$ mF.g^{-1}	GCD test, at current 5.54µA.cm^{-2}	$\sigma_f = 29.1$ MPa, $E_f = 0.3$ GPa	[60]

(*continued*)

Table 1. (*Continued*).

Item number		Configuration of structural supercapacitor			Electrical properties			Mechanical properties	Ref
12	i	CF	DGEBA+$LiClO_4$	FP	Not reported	$C = 5.1$ mF.g^{-1} $E_d = 11.4$ mWh.kg^{-1} $P_d = 47.2$ W.kg^{-1}	CA test, discharging 0.1V for 1800s	$\tau = 5.6$ MPa, $G = 0.8$ GPa	[3]
	ii	Polyaniline-CF	DGEBA+$LiClO_4$	FP		$C = 20.0$ mF.g^{-1} $E_d = 44.6$ mWh.kg^{-1} $P_d = 1.6$ W.kg^{-1}		$\tau = 4.5$ MPa, $G = 1.0$ GPa	
	iii	ACF	DGEBA+$LiClO_4$	FP		$C = 8.4$ mF.g^{-1} $E_d = 18.6$ mWh.kg^{-1} $P_d = 18.4$ W.kg^{-1}		$\tau = 5.1$ MPa, $G = 0.9$ GPa	
	iv	Polyaniline-ACF	DGEBA+$LiClO_4$	FP		$C = 22.2$ mF.g^{-1} $E_d = 49.4$ mWh.kg^{-1} $P_d = 21.6$ W.kg^{-1}		$\tau = 6.3$ MPa, $G = 1.0$ GPa	

(*continued*)

Table 1. (*Continued*).

Item number		Configuration of structural supercapacitor			Electrical properties			Mechanical properties	Ref
13	i	CF	PEGDGE+IL (EMITFSI)	2GF	Not reported	$C = 4.02$ $mF.g^{-1}$ $E_d = 0.006$ $mWh.kg^{-1}$ $P_d = 0.063$ $W.kg^{-1}$	CA test, applying 0.1 V	$\sigma = 160$ MPa, $E = 23.3$ GPa	[45]
	ii	gCF	PEGDGE+IL (EMITFSI)	2GF		$C = 30.82$ $mF.g^{-1}$ $E_d = 0.042$ $mWh.kg^{-1}$ $P_d = 0.456$ $W.kg^{-1}$		$\sigma = 120$ MPa, $E = 19.13$ GPa	
	iii	UgCF	PEGDGE+IL (EMITFSI	2GF		$C = 47.98$ $mF.g^{-1}$ $E_d = 0.067$ $mWh.kg^{-1}$ $P_d =$ $0.788 W.kg^{-1}$		$\sigma = 90$ MPa, $E = 20.72$ GPa	
14		VG-MnO_2-CF	PEGDGE+IL (EMIMTFSI) +LiTFSI	GF	Not reported	$C = 30.7$ $mF.cm^{-2}$ $E_d = 12.2$ $mWh.kg^{-1}$ $P_d = 2.2$ $W.kg^{-1}$	GCD test, at current $0.5 mA.cm^{-2}$ at 0.8V	$\sigma = 86$ MPa, $E = 4.4$ GPa $\sigma_f = 32$ MPa, $E_f = 2.4$ GPa	[44]
15		CF prepreg	Epoxy+PVDF+ LiTf	2GF	Not reported	$C = 11.62$ $mF.g^{-1}$ $P_d = 0.012$ $W.cm^{-2}$	CV test, 0 to 0.4V, scan rate $20 mV.s^{-1}$	$\sigma_f = 47.5$ MPa, $E_f = 8.5$ GPa	[61]

(*continued*)

Table 1. (*Continued*).

Item number		Configuration of structural supercapacitor			Electrical properties			Mechanical properties	Ref
16	i	CF	Epoxy +PEGDGE +IL (EMITSFI) +TiO2	2GF	C = 3.1 mF. g^{-1} E_d = 0.89 mWh.kg^{-1} P_d = 0.17 W. kg^{-1}	Not reported	CA test, discharging 1V for 1000s	σ = 337 MPa, E = 23.7 GPa	[24]
	ii	GNP-Triton X100 -CF	epoxy+PEGDGE +IL (EMITSFI) +TiO_2	2GF	C = 9.6 mF. g^{-1} E_d = 2.86 mWh.kg^{-1} P_d = 1.14 W. kg^{-1}			σ = 294 MPa, E = 24.0 GPa	
17		GA-CF	Epoxy+IL	2DV	C = 56 mF. g^{-1} P_d = 0.02W. kg^{-1}	Not reported	CV test, 0 to 1V, scan rate 1 mV. s^{-1}	Not reported	[51]
18		CAG-CF prepreg	BADEG+ IL (EMIMTFSI)	2GF prepreg	C = 2170 mF.g^{-1} E_d = 1550 mWh.kg^{-1} P_d = 61.7 W. kg^{-1}	C = 1120 mF.g^{-1} E_d = 800 mWh.kg^{-1} P_d = 31.9 W. kg^{-1}	GCD test, at 0.20 mA.cm^{-2} at 2.7 V	Not reported	[43]

(*continued*)

Table 1. (*Continued*).

Item number		Configuration of structural supercapacitor			Electrical properties			Mechanical properties	Ref
19	i	CF	PES +IL ($EMIMBF_4$) +LiTf+ MXene ($Ti_3C_2T_x$)	KF	C = 160 $mF.g^{-1}$ E_d = 20 $mWh.kg^{-1}$ P_d = 1.6 $W.kg^{-1}$	Not reported	CV test, 0 to 0.8V, scan rate 10 $mV.s^{-1}$	σ = 203.0 MPa, E = 15.2 GPa	[62]
	ii	$ZnCuSe_2$ -CF	PES +IL ($EMIMBF_4$) +LiTf+ MXene	KF	C = 8020 $mF.g^{-1}$ E_d = 1110 $mWh.kg^{-1}$ P_d = 26.7 $W.kg^{-1}$			σ = 364.3 MPa, E = 27.4 GPa	
	iii	N-doped $ZnCuSe_2$ -CF	PES +IL ($EMIMBF_4$) +LiTf+ MXene	KF	C = 11930 $mF.g^{-1}$ E_d = 1660 $mWh.kg^{-1}$ P_d = 34.2 $W.kg^{-1}$			σ = 413.1 MPa, E = 33.8 GPa	
	iv	N-doped $ZnCuSe_2$-MXene-CF	PES +IL ($EMIMBF_4$) +LiTf+ MXene	KF	C = 13880 $mF.g^{-1}$ E_d = 1930 $mWh.kg^{-1}$ P_d = 39.2 $W.kg^{-1}$			σ = 454.0 MPa, E = 35.3 GPa	

(*continued*)

Table 1. (*Continued*).

Item number		Configuration of structural supercapacitor			Electrical properties			Mechanical properties	Ref
20		GNP-CF	PEGDGB+TEABF4 +IL ($EMIBF_4$)	no separator	$C = 623\ mF.g^{-1}$ $E_d = 16.9\ mWh.kg^{-1}$ $P_d = 5.2\ W.kg^{-1}$	Not reported	CA test, discharging 1V for 60s	$\sigma = 350$ MPa, $E = 26$ GPa	[63]
21	i	CF	DGEBA+ Epoxy (AG-80) + IL (EMIMTFSI)	2GF	Not reported	$C = 8.7\ mF.g^{-1}$ $E_d = 2.7\ mWh.kg^{-1}$	GCD test, at current 10 $mA.g^{-1}$ at 3V	Not reported	[48]
	ii	Desized CF	DGEBA+ Epoxy (AG-80) + IL (EMIMTFSI)	2GF		$C = 11.9\ mF.g^{-1}$ $E_d = 3.74\ mWh.kg^{-1}$		$\sigma = 268.84$ MPa, $E = 27.53$ GPa $\tau = 19.06$ MPa, $G = 1.56$ GPa	
	iii	AC(15%)-desized CF	DGEBA+ Epoxy (AG-80) + IL (EMIMTFSI)	2GF		$C = 13120\ mF.g^{-1}$ $E_d = 7130\ mWh.kg^{-1}$ $P_d = 232.95\ W.kg^{-1}$		$\sigma = 257.78$ MPa, $E = 23.20$ GPa $\tau = 9.86$ MPa, $G = 0.48$ GPa	

(*continued*)

Table 1. (*Continued*).

Item number		Configuration of structural supercapacitor			Electrical properties			Mechanical properties	Ref
22		CAG-CF	DGEBA+IL (EMIMTFSI)	GF	Not reported	C = 212 $mF.g^{-1}$ E_d = 93 $mWh.kg^{-1}$ P_d = 5.2 $W.kg^{-1}$	GCD test, at current 0.5 $mA.cm^{-2}$	σ = 110 MPa, E = 33 GPa $\tau^{\#}$ = 13.7 MPa, G = 1.7 GPa	[38]
23	i	CF	Epoxy +IL ([Li-G3]TFSI)	2GF	Not reported	C = 19.1 $mF.g^{-1}$	GCD test, at current 0.5 $mA.g^{-1}$	σ_f = 413 MPa, E_f = 21.1 GPa	[64]
	ii	CF-PoPD	Epoxy +IL ([Li-G3]TFSI)	2GF	Not reported	C = 909 $mF.g^{-1}$ E_d = 181.8 $mWh.kg^{-1}$	GCD test, at current 0.5 $mA.g^{-1}$	σ_f = 20.5 MPa, E_f = 3.8 GPa	

2.2 *Multifunctional matrix/ structural electrolyte*

Perhaps the greatest challenge of developing an SSC is the development of a multifunctional matrix/ structural electrolyte which simultaneously acts as the electrolyte which provides ionic conductivity and as the structural matrix which binds the load-bearing fibre electrodes. Therefore, many researchers have given considerable attention to the development of structural electrolytes by using various techniques. In the recent past, many attempts have been devoted to the development of solid and quasi solid-state electrolytes by mixing certain industrial polymers such as epoxy, polyester, and vinyl ester with various additives such as ionic liquids, inorganic salts, and conductive metals. Generally, solid and gel electrolyte-based supercapacitors have a relatively low specific capacitance and low energy density compared to liquid-state electrolytes. However, solid and gel electrolytes have some advantages over liquid electrolytes. For instance, problems such as leakage [2], corrosion, and explosion can be eliminated [65] while the total cell volume is reduced [66].

Among the many instances of possible structural polymer electrolytes, epoxy resin has been studied by many researchers to develop a structural electrolyte for supercapacitors, owing to its high mechanical properties, high thermal and chemical stability, and good adhesive properties [67,68].

Recently, some attempts have been devoted to preparing bicontinuous polymer electrolytes by mixing two polymers that have distinct properties. For example, one rigid polymer (such as DGEBA) and a flexible polymer (such as PEGDGE) are mixed to form the structural polymer electrolyte, in such a way that one phase provides mechanical strength and the other phase supports ion mobility [42].

Generally, epoxy resins are nonconductive. Therefore, ionic liquid or conductive salts are mixed with epoxy resins to imbue the property of ionic conductivity to the polymer matrix. The ionic liquid is a prominent type of electrolyte for blending with polymer matrix when developing blend-type solid polymer electrolytes. Here, the ionic liquid forms a separated phasle inside the porous polymer matrix, so that the transport of ions is not restricted by polymer chains. These electrolytes are mechanically strong and electrochemically and thermally stable, with acceptable levels of conductivity [69]. Very recently, Dharmasiri et al. studied and compared the morphology of an epoxy resin and its solid polymer electrolyte to demonstrate the blending effect of ionic liquid [64]. As illustrated in Figure 9, they have shown that solid polymer electrolyte systems developed by incorporating the ionic liquid into the epoxy resin have a porous or semiporous structure that is large enough to form a network of interconnected channels for ion conductivity. Further, with the higher ionic liquid ratio, ionic liquid droplets align and interact to create many channels for ion conduction.

Besides, Javaid et al. developed a structural electrolyte-based on a PEGDGE/ DGEBA blended polymer, mixed with a lithium salt (LiTFSI). DGEBA is good structural matrix but the high crosslinked density resulted in the hindering of ionic transportation within the polymer matrix. It was demonstrated that the addition of DGEBA into the PEGDGE matrix (optimum ratio was realised as 5: 95) enhanced the mechanical properties of the polymer electrolyte, while the ion transportation activity was compromised [42], as shown in Figure 10.

The mixing of both ionic liquid and conductive salts (for example, lithium based inorganic salts such as LiTFSI, $LiClO_4$, and lithium hexafluorophosphate ($LiPF_6$)) into the epoxy resin is another promising approach currently being investigated to improve

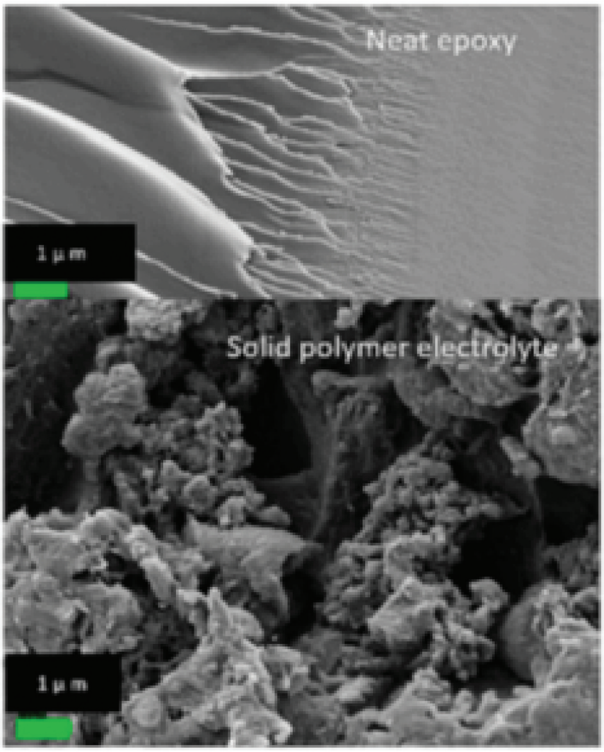

Figure 9. SEM images of neat epoxy and solid polymer electrolyte highlighting the porous morphology [64].

ionic conductivity. To investigate this effect, Shirshova et al. mixed ionic liquid (EMIMTFSI) with lithium salt (LiTFSI) and then prepared three different structural electrolytes by mixing with three commercial epoxy resins in different ratios [70]. They established that lithium salt is essential in obtaining homogeneous samples, as formulations without lithium salt cause phase separation, resulting in a non-homogeneous sample. Further, they provided evidence that the emulsion-based resins had a peak ionic conductivity while showing acceptable mechanical properties. In addition, they found that the composition of the electrolyte greatly affects the properties of the resulting structural electrolytes [.71].

Generally, the neat epoxy has a high mechanical robustness as there are no foreign particles in the epoxy system (see Figure 11a) to hinder the polymer chains. After dissolving ionic liquid in epoxy, the system has comparatively low mechanical strength due to the breakage of some polymer chains. At a low weight ratio of ionic liquid (see Figure 11b), the amount of ionic liquid in the system is not sufficient to form the interconnected channels necessary for ionic conductivity. As illustrated in Figure 11c, with a higher ionic liquid ratio, the number of ion diffusion pathways is large enough to create ionic conductivity. However, when increasing the number of ionic liquid particles inside the epoxy system, mechanical properties drop drastically. When dissolving inorganic salt, as shown in Figure 11d, the

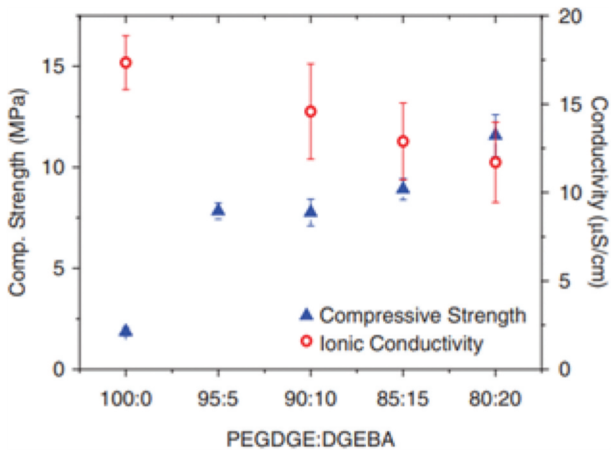

Figure 10. Multifunctionality of PEGDGE polymer electrolyte with increased DGEBA loading: PEGDGE:DGEBA versus compressive strength and ionic conductivity [42].

improved homogeneous microstructure facilitates the creation of a large number of pathways for ionic conductivity, without significantly reducing the mechanical robustness.

Very recently, Qi et al. have highlighted a very important fact about the influence of the weight fraction of the structural electrolyte matrix on the capacitance, energy, and power densities of SSCs [43]. They showed that the reduced structural electrolyte content increases the electrochemical properties, although the mechanical strength declines due to an increased tendency to delaminate. Therefore, it was established that the optimum weight fraction of the structural electrolyte is important for simultaneously high mechanical and electrical properties.

The improvement of ionic conductivity is susceptible to degradation of the mechanical properties due to the conflicting requirements of ion transport and mechanical rigidity. Specifically, mechanical properties like flexural and compression strength, which depend on the matrix, degrade significantly.

In finding solutions for improving the mechanical properties of structural electrolytes, in some studies, researchers have shown that the infusion of nanofillers into the matrix enhances the fracture toughness and flexural strength of the composites [72]. Further, Manoj et al. investigated the effect of carbon-based nanofillers on the properties of the epoxy and showed that the optimum combination of fillers and matrix is necessary for obtaining better results [73]. Besides, Li et al. demonstrated that epoxy resin mixed with mesoporous TiO_2 provides the electrolytes with enhanced electrochemical performance, mechanical strength, and thermal stability [68].

Huang et al. prepared a high-performance structural electrolyte by mixing ionic liquid (EMIMTFSI), polymer matrix (bisphenol F-epichlorohydrin/ trimethylolpropane triglycidyl ether based epoxy resin) and poly(vinyl alcohol) coated carbon nanofibres (PVA-modified CNFs as nanofillers) [74]. The results indicated that PVA-modified CNFs increase the ionic conductivity of the structural electrolyte to 3.18×10^{-2} $S.m^{-1}$, while retaining the Young's modulus at around 800–850 MPa. This improvement in the ionic conductivity of the solid electrolyte is due to the ionic pathways created by

Table 2. Effect of separator thickness for multifunctional performance.

Separator type	Thickness (μm)	Capacitance (nF/m^2)	ILSS (MPa)	E (GPa)	σ_{ult} (MPa)	Ref
Paper 40 g/m^2	71 ± 4	712 ± 118	9.54 ± 0.19	Not reported	Not reported	[75]
Paper 80 g/m^2	89 ± 3	2466 ± 1007	21.76 ± 1.08			
Paper 150 g/m^2	173 ± 5	766 ± 286	36.61 ± 2.34			
Polyacrylate film	50 ± 3	868 ± 198	22.12 ± 3.50			
Polycarbonate film	155 ± 8	206 ± 11	10.19 ± 1.04			
PET film	19 ± 1	1860 ± 1024	20.31 ± 0.58			
PET film	50	447 ± 3.8	29.5 ± 1.3	42.7 ± 3.0	354 ± 66	[76]
PET film	75	300 ± 2.6	30.6 ± 1.7	44.6 ± 0.8	377 ± 15	
PET film	125	193 ± 4.6	32.5 ± 1.4	37.8 ± 4.3	317 ± 36	

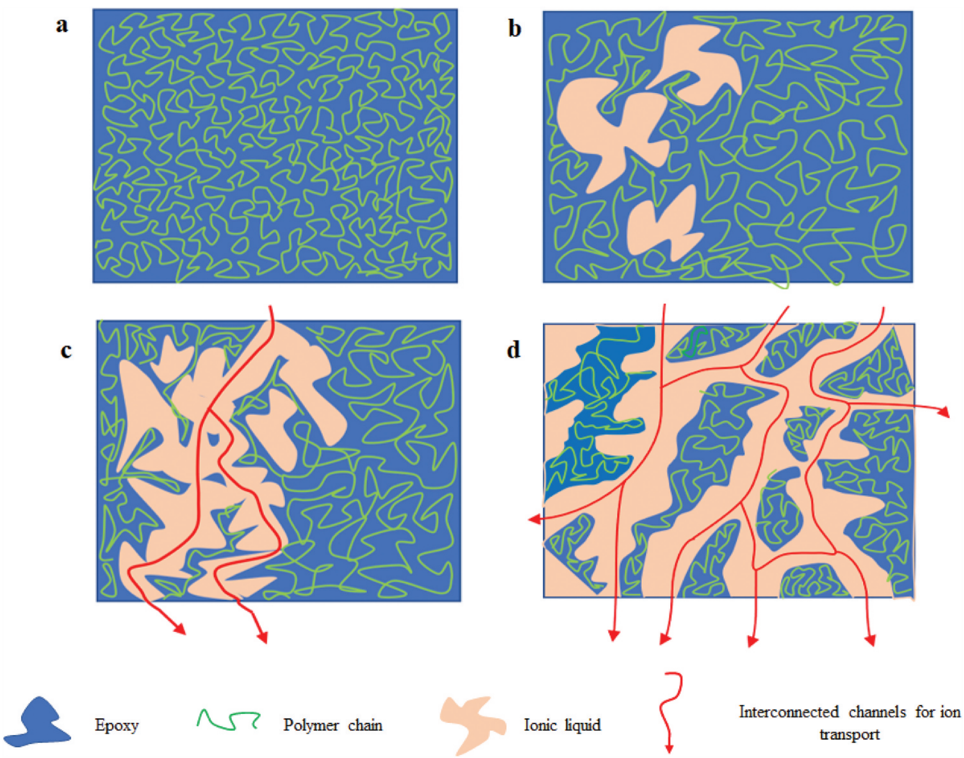

Figure 11. Schematic representation of the formation of interconnected channels in the (a) neat epoxy system, (b-c) epoxy + ionic liquid, (d) epoxy + ionic liquid + Inorganic salt.

CNFs in the epoxy phase. This solid electrolyte has shown a great potential for use in energy storage devices such as SSCs.

In summary, it is clear that the conflict between the mechanical properties and ionic conductivity of structural electrolytes has become a challenging issue in the development of SSCs [17]. Bicontinuous polymer electrolytes mixed with ionic liquid and inorganic salts have attracted great interest in recent years as this has shown comparatively high ionic conductivity. However, the addition of a high proportion of ionic liquid may inversely affect the load-bearing capabilities. In addition, it has been found that the addition of nanofillers in an optimum amount to the structural electrolyte simultaneously enhances the electrical and mechanical properties. Therefore, more research is essential to find the optimum composition of the bicontinuous polymer electrolytes and evaluate suitable materials for infusion into the epoxy mixture to enhance the multifunctional performance of structural electrolytes in terms of ionic conductivity and mechanical robustness.

2.3 *Structural separators*

In the development of SSCs, identifying an appropriate structural separator is challenging. The structural separator should fulfil the primary requirement of high ionic conduction between the electrodes while preventing direct contact between positive and negative electrodes. The interfacial adhesion of the separator to the electrodes is also essential to avoid interlaminar shear failure [32]. On the other hand, the thickness of the separator is a critical factor affecting the supercapacitors' performance as equivalent series resistance (ESR) increases with the separator thickness [17]. Recently, Carlson et al. demonstrated that the separator's multifunctional performance, such as capacitance, dielectric strength, and ILSS, depends on the separator's thickness, as shown in Table 2 [75].

A variety of separators such as glass fibre [24,31], cellulose/filter paper [3,42], ceramic membrane [43], polypropylene membrane [33], geopolymers [29], and kevlar fibre [5,62] have been investigated for SSCs during the past years. Javaid et al. evaluated the performance of SSCs using filter paper (0.18 mm thickness, Whatman Grade 1), polypropylene membrane (PP), and glass fibre fabric as separators [42]. Although polypropylene and cellulose membranes perform well in supercapacitor applications, their inadequate mechanical strength prevents their usage in structural applications. Glass fibre separators (mainly woven materials) have been used for SSCs by many researchers and have shown improved mechanical and electrical properties [33,36,42,77]. In some studies, researchers have mentioned that the surface modification of glass fibre will enhance its electromechanical properties. In particular, grafting CNT or other metal oxide nanofillers enhances the conductivity and compression resistance [32].

Qi et al. compared GF and non-woven polyethylene terephthalate (PET)/ceramic membrane as separators and showed that GF fabric is an effective separator [43]. They adopted a new method which they called structural electrolyte filming to create the structural electrolyte film with targeted thicknesses on the surfaces of the separator. Herein, they suggested two layers of prepreg GF fabrics fully impregnated with the structural electrolyte to achieve improved electronic insulation to prevent local short-circuiting of the SSCs.

Recently, Xu et al. evaluated the performance of a metakaolin-based geopolymer separator which was prepared with metakaolin and different moduli of alkaline activator solution [29]. Geopolymer is an ideal material for the separator due to its dielectric property and high porous structure. As shown in Figure 12, SEM images confirmed the large number of pores in the geopolymer matrix, which provides enough channels for ion storage. The process of geopolymerisation reaction affects the porosity and mechanical property of the geopolymer. A structural supercapacitor with geopolymer separator was proved to exhibit multifunctionality with 33.85 MPa compressive strength and 33.4 $F.g^{-1}$ specific capacitance. However, this improvement of mechanical properties was not as good as that of typical fibre reinforced composites.

In developing the materials for separators, Hubert et al. recently proposed a separator-free structural supercapacitor in which the matrix itself was used to prevent short circuits while it performed its primary task [63]. Partially cured porous matrix film could used for bonding the electrodes. This system showed an increased capacitance and energy density, highlighting the benefit of removing the separator. However, the capability of a low potential window needs to be addressed in future developments of separator-free SSCs as it limits practical applications. Therefore, the choice of separator material has a great impact on the overall performance of SSCs.

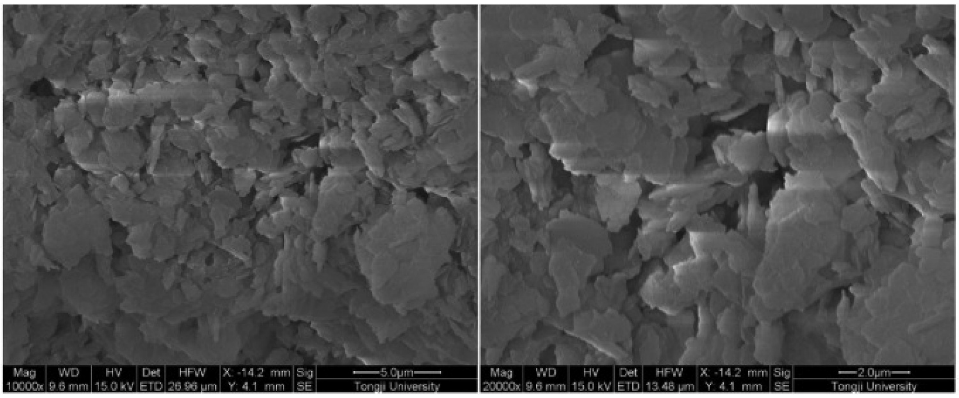

Figure 12. SEM images of metakaolin-based geopolymer [29].

3. Fabrication of SSCs

The fabrication process is critical for achieving the optimal multifunctional properties of the structural supercapacitor composite. In particular, the fabrication process should be able to retain the stiffness, strength, and toughness of structural composites while maximising the proportion of electrochemically active regions [42]. To date, four main approaches have been adopted in fabricating SSCs, namely the brushing method (wet lay-up), resin infusion under flexible tooling (RIFT), the vacuum-assisted resin transfer method (VARTM), and the prepreg method.

In the past, many researchers fabricated SSCs by using the RIFT method [3,33,57] where resin is properly distributed within the composite and reduces the void volume, resulting in a higher fibre volume fraction [3] and an increase in the mechanical properties [33].

In 2014, Javaid and his team developed a structural supercapacitor consisting of woven carbon fibre electrodes, a filter paper (Whatman Grade 1) separator, and a PEGDGE/DGEBA blend polymer electrolyte mixed with lithium salt [41]. The filter paper, was impregnated with polymer electrolyte through the brushing method and then, assembled in a typical configuration where the filter paper was sandwiched between two layers of carbon fibre mat. The composite was laid up by hand, followed by a curing process under an applied pressure of 200 mbar. The same research team used the RIFT method with the support of a vacuum pump (see Figure 13a) for their later work on structural supercapacitor fabrications. They used the RIFT setup after the composite was laid up and allowed the resin to flow into the dry fibre pack loaded into the vacuum-bagged flexible film [57]. In this work, the researchers fabricated the structural supercapacitor using woven carbon fibre electrodes and glass fibre separator impregnated with ionic liquid-based PEGDGE/DGEBA blend polymer electrolyte. They demonstrated that RIFT is an effective method by showing significantly high mechanical and electrical properties for the fabricated structural supercapacitor, compared to the brushing method.

In addition, many researchers have used the VARTM to fabricate SSCs, followed by a curing process [39,48,51,77]. For example, as shown in Figure 13b, Reece et al. used the VARTM setup for the impregnation of the electrolyte-epoxy mixture into the EDLC supercapacitor assembled on a flat glass plate [77]. After the completion of feeding the electrolyte-epoxy mixture, the sample was allowed to cure for 12 h at room temperature while exerting pressure by placing a solid plate of 10 kg on top of the cell assembly. The VARTM is a simple, quick, reliable, and economical method and therefore one of the efficient methods for structural supercapacitor fabrication [39]. However, when using the VARTM, air bubbles may be created in cases of insufficient vacuum and sealing, which will lead to poor mechanical properties. In addition, in cases of an electrolyte mixture with higher viscosity, this method cannot be used.

Nowadays, some researchers have used the prepreg filming process to fabricate SSCs [38,43]. The prepreg method, which is easily scaled up in industry, is increasingly popular among the research community as the way of realising high-performance SSCs. As discussed in Section 2.3, Qi and his team suggested the resin filming process to fabricate the structural supercapacitor via the sandwiching of two CAG loaded carbon fibre partial prepregs with the separator prepreg [43]. Ionic liquid-based structural electrolyte which contains bisphenol A diglycidyl ether (BADGE) epoxy, isophorone diamine (IPDA) hardener, and ionic liquid (EMIMTFSI) was applied onto the inner face of the CAG–carbon fibre electrodes to form carbon fibre partial prepregs. An adjustable height film applicator was used to make the structural electrolyte films with the required thicknesses. The prepreg stack of carbon fibre/separator/carbon fibre was laid up for the curing process after copper adhesive strips were attached to the outer faces of the CAG–carbon fibre electrodes. In this way, the weight fraction of the structural electrolyte can be properly controlled, although minor differences can be observed between replicates. Therefore, nominally identical devices can be manufactured by this method, ensuring the repeatability of device fabrication.

As discussed in previous sections, in much of the research epoxy-based solid electrolytes have been used when fabricating SSCs. However, solid electrolyte-based supercapacitors have relatively low specific capacitance and energy density compared to liquid electrolyte-based supercapacitors. In order to address this issue, Reece et al. recently proposed a novel design method and fabricated a liquid electrolyte-based supercapacitor composite with structural properties [10]. This is an attempt to implement the structural supercapacitor in a honeycomb structure typically used in a sandwich composite. As shown in Figure 14, liquid organic electrolyte (1 M $TEABF_4$) based supercapacitors were embedded in the skins and integrated into the honeycomb core where the aluminium faces of the core behave as the current collector. This is a good initiative to deviate from the typical supercapacitor array design and fabricate the SSCs by adapting the well-developed architecture of fibre reinforced

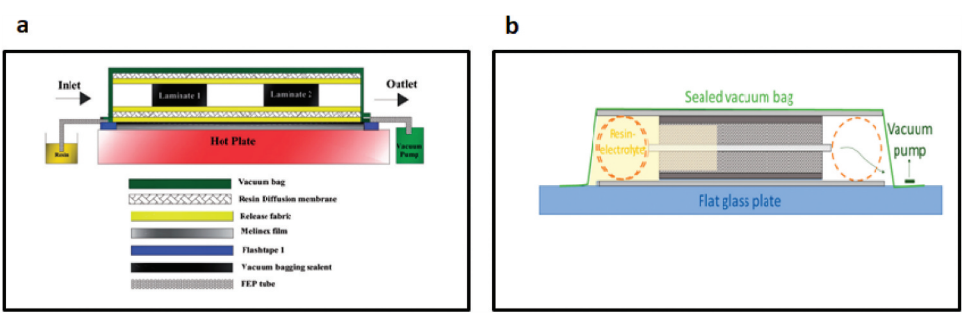

Figure 13. Schematic of (a) the RIFT process [57], (b) the VARTM for the impregnation of the structural electrolyte into EDLC mounted on a flat glass plate [77].

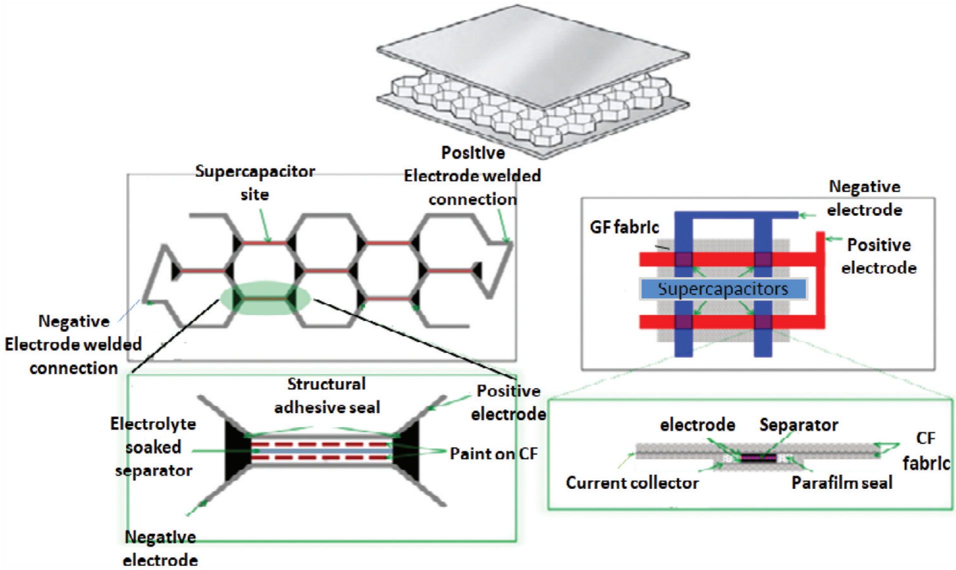

Figure 14. Conceptual diagram of the supercapacitor functional composite with structural properties: (a) sandwich composite structure, (b),(c): supercapacitor functional honeycomb core; (d), (e) supercapacitor functional skin; (b) and (d) plane-view of overall designs, (c) and (e) detail depicting a supercapacitor device for the honeycomb core (c) and the skin (e) [10].

composites. However, there have not been sufficient successful attempts to demonstrate the innovative geometrical design for SSCs, so far.

Recently, Sun et al. [78] fabricated a hybrid laminated structural composite (see Figure 15) to simultaneously improve the mechanical and electrical properties. The outer layer consists of four sandwiched kevlar fabric/epoxy prepregs and the inner layer is a thin interleaf of carbon fibre/solid electrolyte supercapacitor.

The developed model exhibited high electrochemical and mechanical performance. The specific capacitance, energy density and power density values which were

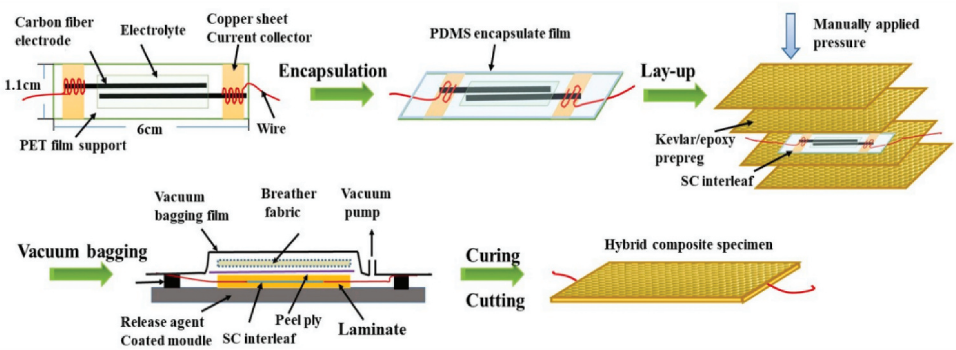

Figure 15. Fabrication process of the Kevlar/SC/epoxy hybrid composite [78].

normalised to the total hybrid composite weight, were 0.872 $mF.g^{-1}$, 0.08 $mWh.kg^{-1}$, and 9.2 $mW.kg^{-1}$ respectively at current density of 0.05 $A.cm^{-3}$. The hybrid laminate flexural strength and flexural modulus were measured as 192 MPa and 9.3 GPa, respectively. Moreover, the impressive characteristic of this panel is its capability for working in variable operating voltages. This is done by changing the number of supercapacitor interleaves embedded in the hybrid laminate or by changing the connection of supercapacitor interleaves (parallel or series, see Figure 16a). For instance, in the series connection, the operating voltage which was initially at 0.8 V increased to 2.4 V with nearly identical discharge, and the discharge time at 0.8 V was increased by three times in parallel connection, as shown in Figure 16b. Therefore, interleaving the supercapacitors into the fibre reinforced laminates is a prominent method to produce successful SSCs with well-balanced mechanical and electrochemical properties. However, delamination cracks between the supercapacitor interleaf and kevlar/epoxy lamina were initiated when the external load increased to a certain level, due to the material inhomogeneity-induced stress concentration.

Besides, Mapleback et al. proposed a new design methodology for SSCs to eliminate the poor ionic conductivity due to solid electrolytes [79]. They proposed a structure where the supercapacitor electrolyte was localised within the composite structure to maintain high electrochemical properties in localised areas and high mechanical performance elsewhere.

In summary, it was observed that many researchers have discussed the major challenge of SSCs in achieving high mechanical and electrochemical properties simultaneously. Particularly, the mechanical and electrical performance of a structural energy storage device is dependent on the fibre/matrix interaction. The ion's ability to reach the fibre surface is crucially important for high electrochemical properties. However, poor interfacial load transmission results, due to the direct access of ions to the fibres. Therefore, SSCs based on solid electrolyte are limited in both properties to a certain level due to the conflicting requirements of ion transport and mechanical rigidity, which results in a low-performance device. When carefully studying the recent approaches, a promising one is the

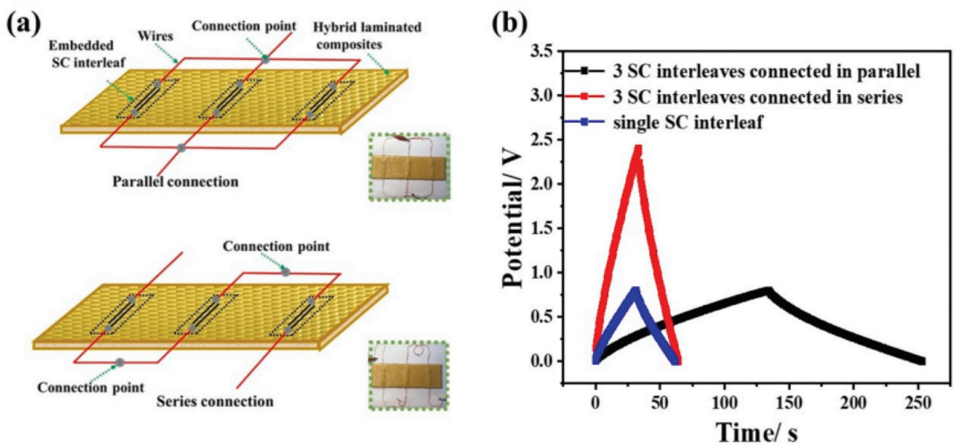

Figure 16. Schematics and photographs of three SC interleaves embedded in a hybrid laminated composite, connected in parallel and in series, (b) GCD curves of the hybrid composites with single SC interleaf, and with three SC interleaves connected in parallel and in series [78].

sandwich composite panel with supercapacitor functional core layers covered by outer layers with higher structural properties. Although the core layers might have minimum structural properties, they will be protected by the outer layers from the external forces. Herein, the individual properties can be maximised independently, and finally, higher overall performance can be achieved by the developed hybrid laminate. However, in the integration of supercapacitors into high-performance carbon fibre composites, the materials used for the supercapacitor are required to be compatible with high temperature and pressure. Also, to protect the supercapacitor from the infusion of epoxy resin, encapsulation polymer layers need to be used on the top and bottom.

4. Performance evaluation of SSCs

The ultimate goal of a structural supercapacitor is to enhance the multifunctionality of the developed device, in particular balancing the mechanical and electrical functionalities without substantially sacrificing one property over the other. Therefore, the evaluation of multifunctional performance is essential to identify the best SSCs. Notably, in much of the research, when presenting the multifunctional performance, the mechanical properties and electrochemical properties have been reported independently for the developed structural composite. To evaluate the mechanical performance, tensile, compressive, three-point bending, and interlaminar shear tests have been investigated. The mechanical properties of different SSCs have thus been compared using the results of the above testing.

On the other hand, many researchers have widely used testing such as cyclic voltammetry (CV), galvanostatic charge-discharge (GCD), choroamperometric test (CA), and electrochemical impedance spectroscopy (ElS) for electrochemical analysis. However, a standard approach for presenting the electrochemical performance is still lacking and thus, the comparison of different structural supercapacitors in terms of energy storage is challenging [76]. For instance, when presenting electrochemical data, some researchers present the results of the three electrode system and other present the results of the two electrode system. In addition, the voltages and type of testing used for characterisation are sensitive to characteristics like energy and power density. Moreover, some researchers normalise the electrochemical data with respect to the active material mass while others normalise with respect to the device mass. Therefore, in the future, a consistent method or acceptable standard protocol is obviously required to enable comparison and, consequently, to identify the most promising structural supercapacitor in terms of energy storage when scrutinising the literature.

It is well known that independent evaluations are not adequate for evaluating the multifunctional performance of SSCs. Thus, nowadays, researchers have used different approaches to evaluate multifunctional performance. In demonstrating such an approach, Carlson et al. measured the multifunctionality of energy storage structural devices by evaluating the energy density with respect to specific interlaminar shear strength [71]. Consequently, this method enables us to assess the applicability of these structures to store energy and evaluate their potential to reduce system weight.

Examples of other ways to evaluate multifunctionality are that Ganguly et al. correlated device energy efficiency against mechanical elastic modulus efficiency [44], while Shirshova et al. correlated specific capacitance against compressive modulus [31].

For practical applications, SSCs need to bear mechanical loads and store energy simultaneously. Thus, nowadays, some researchers characterise the electrochemical performance under mechanical loading as this represents the real situation and is more meaningful for evaluating the performance of SSCs. For instance, Zhou et al. evaluated the electrochemical performance of their developed SSC under external mechanical loads (three-point bending and tensile test), as shown in Figures 17a and 17b. According to their results (see Figures 17c and 17d) they found that under different loads, no significant difference in CV curves was observed. Also the electrical connection was preserved until the whole structure started to fail mechanically. This implies that the composite structure provides a safety guarantee for flexible energy storage devices, which is an appealing feature for practical applications.

In addition, O'Brien et al. [80] developed an interesting approach recently to evaluate multifunctional efficiency (η_{mf}) which calculates the weight saving achieved by the multifunctional composite compared to its baseline. To achieve weight saving, the following criteria should be satisfied:

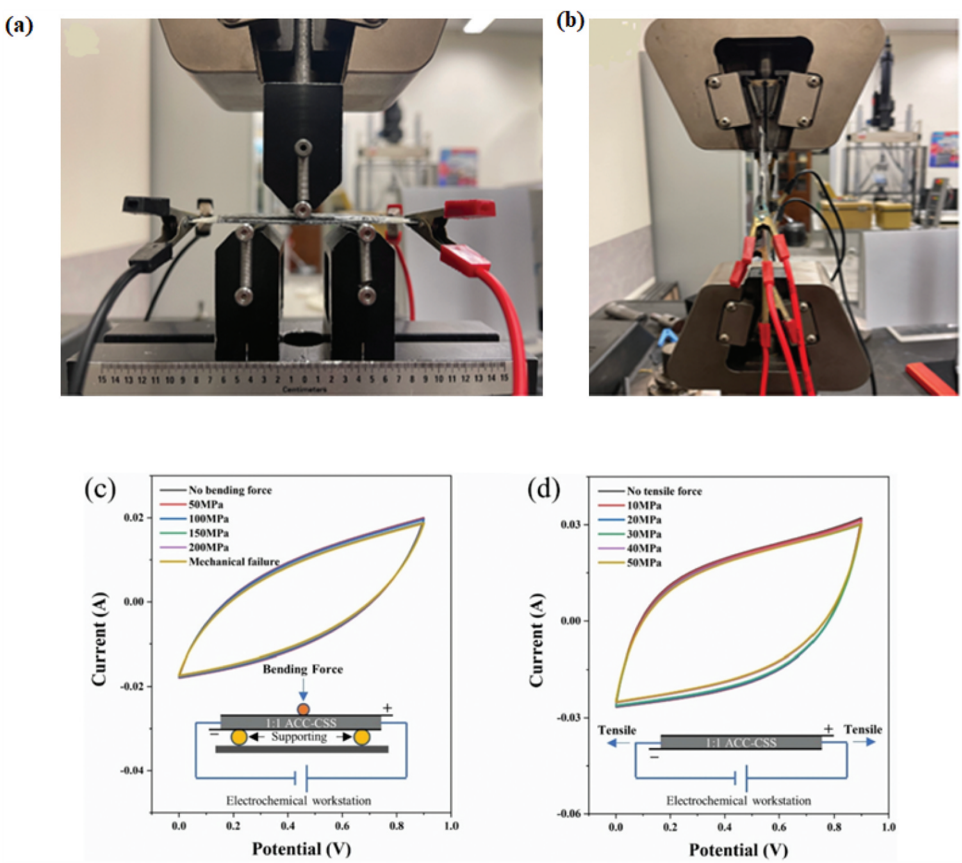

Figure 17. Image of (a) in-situ 3-point bending-electrochemical measurement, (b) in-situ tensile-electrochemical measurement, (c) CV test under three-point bending test, (d) CV test under tensile test [81] [].

$$\eta_{mf} = \eta_e + \eta_s > 1$$

structural efficiency $(\eta_s) = \frac{E_{mf}}{E}$

Capacitive energy efficiency $(\eta_e) = \frac{T_{mf}}{T}$

where T_{mf} and E_{mf} are the energy density and specific stiffness of the multifunctional composite, respectively. T and E represent the specific energy of a monofunctional supercapacitor and the specific stiffness of a monofunctional laminate, respectively.

5. Application prospects of structural supercapacitors

Structural supercapacitors are suited to a wide range of applications where both load-bearing and energy storage functions are required simultaneously. For a long time now, CFRP materials have typically been dominant in many engineering fields as an alternative material for metallic structures. The new development of CF-based SSCs will widen CFRPs' application prospects due to their capability of providing not only mechanical support to the structures but also energy storage. Particularly, such load-bearing structures can work as devices that store and deliver electricity from renewable energy sources. Although, this has not yet reached the level needed for real-world utilisation, many researchers have demonstrated its suitability for various applications. In particular, such designs are suitable for applications such as parts of smart building bodies (wall bricks and roof tiles), body parts of energy generation units, body parts of electric and hybrid vehicles (the door panels, roof, chassis, and bonnet), body parts of electric rails, the structure of unmanned aerial vehicles (satellites and drones), the

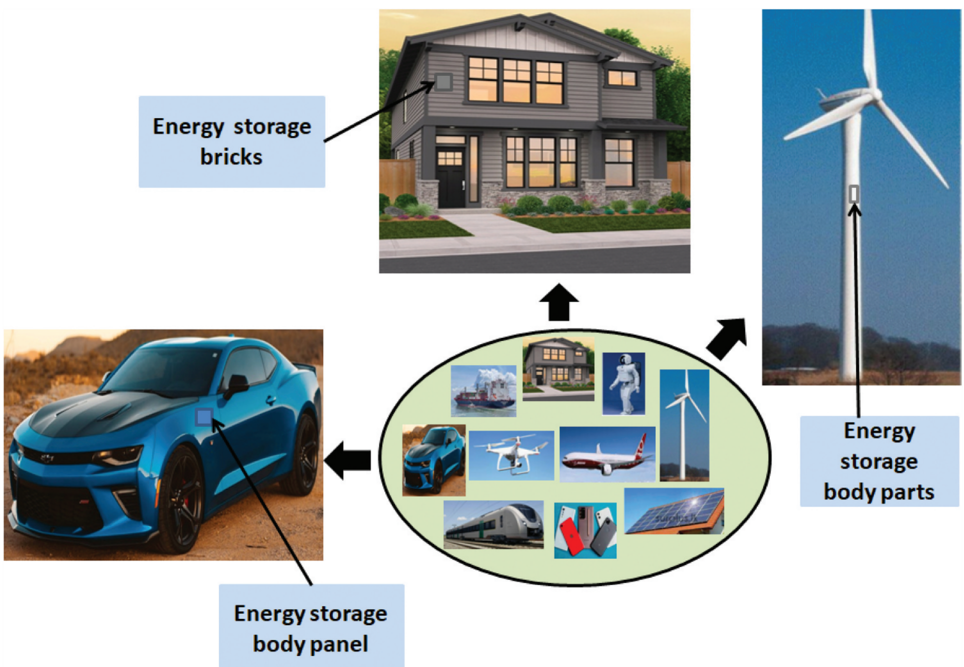

Figure 18. Application prospects of structural supercapacitors.

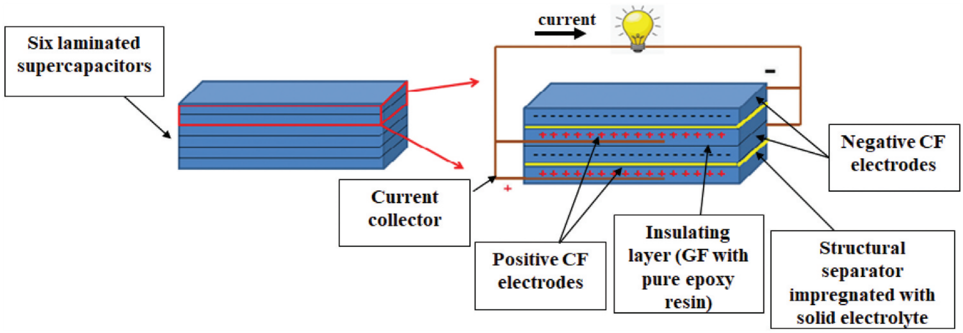

Figure 19. Proposed multilayer concept for supercapacitor functional panel.

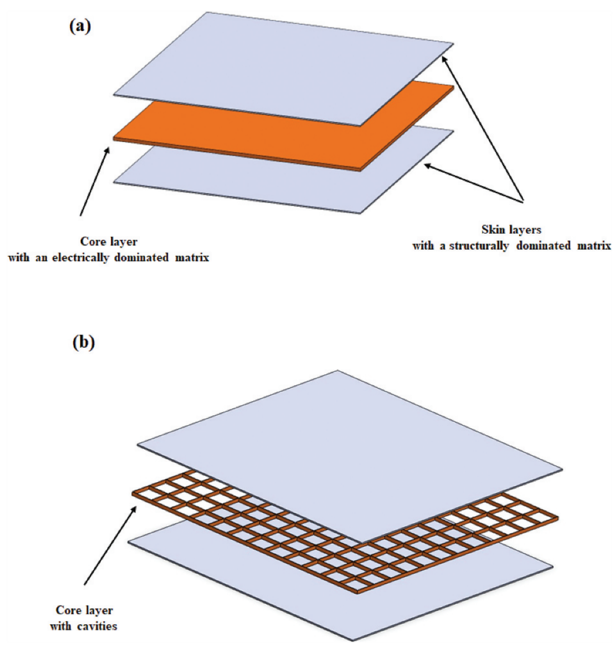

Figure 20. Proposed sandwich structure for SSCs.

structure of mobile robots, and beyond (see Figure 18). These developments are useful in cases where access to the national grid is not feasible and the only available source of energy is renewable energy resources such as wind or solar. In such a situation, body parts can store the electricity when the source is available and use the stored electricity when the source does not produce enough power.

Moreover, substantial weight saving can be achieved from this concept by reducing parasitic materials in structures. In particular, this method offers a number of design opportunities for applications such as aircraft, spacecraft, ships, drones, satellites, and tangential applications where weight is a critical factor. In addition, as shown in Figure 18, power generation units can store the generated electricity in their structural parts without an external battery system, when the parts have been made with energy storage capability. For instance, wind towers and solar panels can store generated electricity in their supporting structural parts for later use.

Although a wide range of application prospects exists for SSCs, no significant work so far has been highlighted for structural supercapacitor integrated structure. Therefore, more efforts are required in future to revolutionise SSCs to be used in many practical applications.

6. Challenges and future development

The major challenge of structural supercapacitors is to fabricate a supercapacitor functional composite while imparting the necessary mechanical properties [37]. The whole device should be able to withstand mechanical stresses from compression, delamination, and fatigue due to real life operational loads. Also, it should tolerate environmental conditions such as temperature extremes and fire resistance while sustaining electrochemical properties such as high energy and power densities.

In the past decade, research interest in developing multifunctional composites with CF reinforcement for storing energy has increased significantly. However, the main electrochemical challenge faced by SSCs is the low capacitance due to the low surface area of the CF electrodes and low ionic conductivity due to the low ionic diffusivity of the solid electrolyte. In order to address these two issues, a range of different techniques has been studied by many researchers, such as the activation of CF fabrics, the development of various types of structural electrolytes with improved properties, the development of compatible separators, and new architectural designs when fabricating the structural supercapacitor.

Surface modification of the CF fabric is considered to be the best approach to enhance the electrochemical properties of structural supercapacitors. Within this approach, the most promising is the surface grafting of CAG, CNTs, graphene, or oxidising materials onto the carbon fibres. However, in some cases, surface modifications could lead to inverse results for the overall performance of SSCs. Therefore, it is clear that the surface modification of CF fabrics creates a new research direction when developing electrodes for SSCs.

It has been shown that solid-state electrolytes have a substantial potential for SSCs. However, as solid-state electrolytes have a relatively low ionic conductivity compared to liquid-state electrolytes, more research is needed to focus on the formulations of multifunctional electrolytes, aiming at enhancing the ion movements in the electrolytes without significantly degrading their mechanical properties. For instance, it is essential to identify the optimum composition of ionic liquids in the polymer matrices in order to optimise the mechanical and electrical performance of structural supercapacitors [17]. In addition, recent studies have confirmed that the mixing of nanofillers simultaneously improves the electrochemical and mechanical properties. However, finding suitable nanofillers in the optimum amount is challenging. Therefore, this opens new avenues for future research.

Moreover, the separators used for SSCs would have a substantial influence on the overall performance of the device although less attention has been given to improving separators' properties so far. Some researchers have shown an improved overall performance of a structural supercapacitor which does not have a separator; however, these investigations are still in the early stages. Therefore, in the future, more research is required to identify suitable matrices which can eliminate the requirement for separators and the capability to prevent short-circuiting. SSCs have commonly used GF as the insulating separator, though some characteristics of GF are unfavourable for SSCs' performance. Therefore, it is desirable to develop new materials for structural separators and new surface modification techniques to improve the performance of separators.

Since the current performance of structural supercapacitors is far from realisation for pertinent engineering applications owing to low energy and mechanical properties, new design architectures should be investigated. Insufficient effort has been devoted to the development of structural composites consisted with bank of supercapacitors. By scrutinising the various configurations found in the referred literature, in this article, the authors could conceptually propose a new multilayer design for developing a high-performance supercapacitor functional panel with the use of fabricating methods for conventional fibre reinforced composites, as shown in Figure 19. Each carbon fibre layer alternatively behaves as positive and negative electrodes. Subsequently, the whole composite acts as a panel made of a bank of supercapacitor functional layers. These supercapacitor functional layers can be electrically connected, either in series mode or parallel mode, to adjust the overall operating voltage and capacitance of the panel. This kind of design will facilitate the easy customisation of electrochemical and mechanical properties to ensure their compatibility with every single application.

Alternatively, the sandwich structure, as shown in Figure 20a, could be a promising approach to simultaneously maximise mechanical and electrical properties. Here, the structural supercapacitor core can be designed with an electrically dominated matrix and structural laminates (skin layers) can be designed with a structurally dominated matrix. In addition, this approach is interesting due to its capability to replace the structural electrolyte used in the core layers with pure ionic liquid and its capability to enclose the core layers to avoid electrolyte leakage.

Electrochemical performance typically depends on the size of the supercapacitor and small size supercapacitors can perform well. Thus, this architecture can be modified with cavities inside the core layer, as shown in Figure 20b, and small supercapacitors can be embedded into the cavities to improve the overall performance. The number of supercapacitors and number of skin layers could be customised according to the expected energy output and mechanical properties. Moreover, the skin layers can be designed to tolerate extreme environmental conditions, such as high temperatures and fire resistance, in such a way that the developed composite can be utilised for high-end engineering applications. Studying and developing this concept would be a promising aspect for future researchers.

Apart from this, the reduction of the overall cost of structural supercapacitor fabrication is another important aspect although it has not yet received much attention. Thus, adequate consideration should be given to this in developing new materials and new fabrication techniques for SSCs.

7. Conclusion

In this article, the authors have comprehensively and critically discussed the novel concept of SSCs based on CF reinforced polymers that could be used for many engineering applications in the civil and defence industries, e-mobilty sector, smart electronic devices (healthcare devices, sports equipment, artificial intelligence), and smart grids. An emerging interest was seen among researchers in the development of supercapacitor functional composites in recent years because of their intermediate power and energy densities. In addition, compared to other energy storage devices, supercapacitors are superior in terms of charge-discharge rate, cyclability, and safe operation. Structural supercapacitors with laminated architecture are easy to integrate into fibre reinforced polymeric components, making them more suited for structural power applications.

In this article, the recent development of structural supercapacitors was summarised and critically discussed, with the focus on the surface modification of CF electrodes, formulations of multifunctional electrolytes, and the development of insulating separators. In particular, various approaches were presented to improve the specific surface area and the specific capacitance of carbon fibres without affecting their structural properties. The development of polymer matrix as the structural electrolyte, the most challenging aspect of SSCs, was among the focal points of this review as the ionic conductivity of the structural supercapacitor mainly depends on the conductivity of the electrolyte.

Many researchers have pointed out that a mechanical electrochemical trade-off is often unavoidable when optimising the multifunctional performance of structural electrolytes. In order to address this issue, the combination of ionic liquids, inorganic salts, and polymer blends, which has been discussed by many researchers, was summarised. The fabrication processes and novel designs for future development were also discussed in detail in the latter part of the article.

As a promising novel design, a sandwich composite structure with supercapacitor functional core and structurally strong skins was proposed. The benefits of this include that many structural properties can be incorporated into the skins and electrical properties are improved in the core layer. In the future, this kind of design will avoid the main obstacle of the mechanical electrochemical trade-off.

Despite the rapid development of structural supercapacitors, to develop high-performance structural supercapacitors, numerous challenges remain to be tackled and taken into consideration. It is essential to pay attention to the operating conditions of structural supercapacitors since they involve exposure to various in-service conditions. In this regard, structural supercapacitors should be strong enough to tolerate temperature extremes, fire resistance, impact, and damage. Besides, in the future, structural supercapacitors may revolutionise many applications in civil and defence industries, the e-mobilty sector, smart electronic devices, and smart grids. However, there has not been a significant progressive development of researched supercapacitors that are cited in this review. Therefore, there is still plenty of room for new developments of application-oriented structural supercapacitors. With this study, the authors expect to give useful insights into the challenges that need to be overcome for the development of high-performance structural supercapacitors in real life applications.

Nomenclature

EDLC	Electric double layer capacitors
CFRPs	Carbon fibre reinforced polymers
CNTs	Carbon nanotubes
SSCs	Structural supercapacitors
CF	Carbon fibre
GF	Glass fibre
MnO_2	Manganese dioxide

NiO	Nickel oxide
TiO_2	Titanium dioxide
CAG	Carbon aerogel
GO	Graphene oxide
GNPs	Graphene nanoplatelets
GA	Graphene aerogel
EMITFSI	1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl) imide
LiTFSI	Lithium bis(trifluoromethane) sulfonimide
PES	Polyester resin
$EMIMBF_4$	1-ethyl-3-methylimidazolium terafluoroborate
LiTf	Lithium trifluoromethanesulfonate
DGEBA	Diglycidylether of bisphenol-A
SEM	Scanning electron microscopic
PAni	Polyaniline
CA	Chronoamperometry test
GCD	Galvanostatic charge-discharge
CV	Cyclic voltammetry
$LiClO_4$	Lithium perchlorate
PC	Propylene carbonate
RIFT	Resin infusion under flexible tooling
PEGDGE	Poly(ethylene glycol) diglycidylether
C	Specific capacitance
E_d	Energy density
P_d	Power density
τ	Shear strength
G	Shear modulus
τ^*	Shear strength at 0.5% shear- strain
σ	Tensile strength
E	Young's modulus
σ_f	Flexural strength
E_f	Flexural modulus
IL	Ionic liquid
2GF	2 pieces of glass fibre fabric
PP	Polypropylene membrane
MSP	Mesoporous silica particles
ACF	Activated carbon fibre
FP	Filter paper
KF	Kevlar fibre
CP	Cellulose paper
$TEABF_4$	Tetraethylammonium tetrafluoroborate
gCF	Graphene nanoflake-coated carbon fibre
UgCF	Urea activated gCF
VG	Vertical graphene
PVDF	Polyvinylidene fluoride
2DV	2 layers of dielectric electrospun veil separator
EMIMTFSI	1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide
EMIBF4	1-ethyl-3-methylimidazolium tetrafluoroborate
AC	Activated carbon
$\tau^{\#}$	Shear strength at 1% strain
$LiPF_6$	Lithium hexafluorophosphate
PE	Polythene
CNFs	Carbon nanofibres
PoPD	poly(o-phenylenediamine)
SC	Supercapacitor
ESR	Equivalent series resistance
PET	polyethylene terephthalate

VARTM	Vacuum-assisted resin transfer method
BADGE	Bisphenol A diglycidyl ether

Disclosure statement

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 article.

Funding

This research was supported by the AHEAD operation (Accelerate Higher Education Expansion and Development), a World Bank funded project in Sri Lanka.

ORCID

Jayani Anurangi http://orcid.org/0000-0002-7685-9845
Madhubhashitha Herath http://orcid.org/0000-0002-6796-0802
Dona T.L. Galhena http://orcid.org/0000-0002-8619-164X
Jayantha Epaarachchi http://orcid.org/0000-0001-6472-9405

Data availability statement

Previously published data has been used in this review article. The raw data required to reproduce these findings are available to download from the original publisher or authors of each reference.

References

[1] Libich J, Máca J, Vondrák J, et al. Supercapacitors: properties and applications. J Energy Storage. 2018;17:224–227.
[2] Zhong C, et al. A review of electrolyte materials and compositions for electrochemical supercapacitors. Chem Soc Rev. 2015;44(21):7484–7539.
[3] Javaid A, et al. Multifunctional structural supercapacitors based on polyaniline deposited carbon fiber reinforced epoxy composites. J Energy Storage. 2021;33:102168.
[4] Liu C, et al. Graphene-based supercapacitor with an ultrahigh energy density. Nano Lett. 2010;10(12):4863–4868.
[5] Deka BK, et al. Bimetallic copper cobalt selenide nanowire-anchored woven carbon fiber-based structural supercapacitors. Chem Eng J. 2019;355:551–559.
[6] Salkuti SR. Energy storage technologies for smart grid: a comprehensive review. Majlesi J Electr Eng. 2020;14(1):39–48.
[7] Bae S-H, et al. Load-bearing supercapacitor based on bicontinuous PEO-b-P(S-co-DVB) structural electrolyte integrated with conductive nanowire-carbon fiber electrodes. Carbon. 2018;139:10–20.
[8] Lee JH, et al. Restacking-inhibited 3D reduced graphene oxide for high performance supercapacitor electrodes. ACS Nano. 2013;7(10):9366–9374.
[9] Ladpli P, et al. Multifunctional energy storage composite structures with embedded lithium-ion batteries. J Power Sources. 2019;414:517–529.
[10] Reece R, Lekakou C, Smith PA. A high-performance structural supercapacitor. ACS Appl Mater Interfaces. 2020;12(23):25683–25692.
[11] Berjoza D, Jurgena I. Influence of batteries weight on electric automobile performance. Eng for Rural Dev. 2017;16:1388–1394.
[12] Adam TJ, et al. Multifunctional composites for future energy storage in aerospace structures. Energies. 2018;11(2).

[13] Galos J, Best AS, Mouritz AP. Multifunctional sandwich composites containing embedded lithium-ion polymer batteries under bending loads. Mater Des. 2020;185:108228.
[14] Pattarakunnan K, Galos J, Das R, et al. Tensile properties of multifunctional composites embedded with lithium-ion polymer batteries. Compos Part A Appl Sci Manuf. 2020;136:105966.
[15] Ladpli P, et al. *Design of multifunctional structural batteries with health monitoring capabilities*. in *Proceedings of the 8th European workshop on structural health monitoring (EWSHM 2016)*. Bilbao Spain; 2016.
[16] Pereira T, Guo Z, Nieh S, et al. Energy storage structural composites: a review. J Compos Mater. 2009;43(5):549–560.
[17] Chan K-Y, Jia B, Lin H, et al. A critical review on multifunctional composites as structural capacitors for energy storage. Compos Struct. 2018;188:126–142.
[18] Hudak NS, Schlichting AD, Eisenbeiser K. Structural supercapacitors with enhanced performance using carbon nanotubes and polyaniline. J Electrochem Soc. 2017;164(4):A691–A700.
[19] Attar P, Galos J, Best AS, et al. Compression properties of multifunctional composite structures with embedded lithium-ion polymer batteries. Compos Struct. 2020;237:111937.
[20] Xiao Y, et al. The effect of embedded devices on structural integrity of composite laminates. Compos Struct. 2016;153:21–29.
[21] Pakdel E, et al. Recent progress in recycling carbon fibre reinforced composites and dry carbon fibre wastes. ResouConserv Recycl. 2021;166:105340.
[22] Xie S, et al. Recent advances toward achieving high-performance carbon-fiber materials for supercapacitors. ChemElectroChem. 2018;5(4):571–582.
[23] Bigdeloo M, et al. Review on innovative sustainable nanomaterials to enhance the performance of supercapacitors. J Energy Storage. 2021;37:102474.
[24] Sánchez-Romate XF, et al. A proof of concept of a structural supercapacitor made of graphene coated woven carbon fibers: EIS study and mechanical performance. Electrochim Acta. 2021;370:137746.
[25] Muralidharan N, et al. Carbon nanotube reinforced structural composite supercapacitor. Sci Rep. 2018;8(1):17662.
[26] Fang C, Zhang D. High multifunctional performance structural supercapacitor with Polyethylene oxide cement electrolyte and reduced graphene oxide@CuCo2O4 nanowires. Electrochim Acta. 2022;401:139491.
[27] Chung DDL. A review of multifunctional polymer-matrix structural composites. Compos Part B Eng. 2019;160:644–660.
[28] Asp LE, Greenhalgh ES. Structural power composites. Compos Sci Technol. 2014;101:41–61.
[29] Xu J, Zhang D. Multifunctional structural supercapacitor based on graphene and geopolymer. Electrochim Acta. 2017;224:105–112.
[30] Fang C, Zhang D. High areal energy density structural supercapacitor assembled with polymer cement electrolyte. Chem Eng J. 2021;426:130793.
[31] Shirshova N, et al. Structural composite supercapacitors. Compos Part A Appl Sci Manuf. 2013;46:96–107.
[32] Deka BK, et al. Recent development and challenges of multifunctional structural supercapacitors for automotive industries. Int J Energy Res. 2017;41(10):1397–1411.
[33] Qian H, et al. Multifunctional structural supercapacitor composites based on carbon aerogel modified high performance carbon fiber fabric. ACS Appl Mater Interfaces. 2013;5(13):6113–6122.
[34] Artigas-Arnaudas J, et al. Surface modifications of carbon fiber electrodes for structural supercapacitors. Appl Compos Mater. 2021.
[35] Gaikwad N, et al. Advanced polymer-based materials and mesoscale models to enhance the performance of multifunctional supercapacitors. J Energy Storage. 2023;58:106337.
[36] Xu Y, et al. Structural supercapacitor composites: a review. Compos Sci Technol. 2021;204:108636.
[37] Zhou H, et al. Structural composite energy storage devices — a review. Mater Today Energy. 2022;24:100924.
[38] Pernice MF, et al. Mechanical, electrochemical and multifunctional performance of a CFRP/carbon aerogel structural supercapacitor and its corresponding monofunctional equivalents. Multifunct Mater. 2022;5(2):025002.

[39] Pal B, Yang S, Ramesh S, Thangadurai V and Jose R. (2019). Electrolyte selection for supercapacitive devices: a critical review. Nanoscale Adv., 1(10), 3807–3835. 10.1039/C9NA00374F
[40] Deka BK, et al. Multifunctional CuO nanowire embodied structural supercapacitor based on woven carbon fiber/ionic liquid–polyester resin. Compos Part A Appl Sci Manuf. 2016;87:256–262.
[41] Javaid A, Irfan M. Multifunctional structural supercapacitors based on graphene nanoplatelets/carbon aerogel composite coated carbon fiber electrodes. Mater Res Express. 2018;6(1):016310.
[42] Javaid A, et al. Multifunctional structural supercapacitors for electrical energy storage applications. J Compos Mater. 2013;48(12):1409–1416.
[43] Qi G, et al. The influence of fabrication parameters on the electrochemical performance of multifunctional structural supercapacitors. Multifunct Mater. 2021;4(3):034001.
[44] Sha Z, et al. Synergies of vertical graphene and manganese dioxide in enhancing the energy density of carbon fibre-based structural supercapacitors. Compos Sci Technol. 2021;201:108568.
[45] Ganguly A, et al. Multifunctional structural supercapacitor based on urea-activated graphene nanoflakes directly grown on carbon fiber electrodes. ACS Appl Energy Mater. 2020;3(5):4245–4254.
[46] Qian H, et al. Activation of structural carbon fibres for potential applications in multifunctional structural supercapacitors. J Colloid Interface Sci. 2013;395:241–248.
[47] Chung DDL. Development, design and applications of structural capacitors. Appl Energy. 2018;231:89–101.
[48] Ding Y, et al. High-performance multifunctional structural supercapacitors based on in situ and ex situ activated-carbon-coated carbon fiber electrodes. American Chemical Society: Energy & Fuels; 2022.
[49] Samsur R, et al. Fabrication of carbon nanotubes grown woven carbon fiber/epoxy composites and their electrical and mechanical properties. J Appl Phys. 2013;113(21):214903.
[50] Nguyen S, et al. Mechanical and physical performance of carbon aerogel reinforced carbon fibre hierarchical composites. Compos Sci Technol. 2019;182:107720.
[51] Subhani K, et al. Graphene aerogel modified carbon fiber reinforced composite structural supercapacitors. Compos Commun. 2021;24:100663.
[52] Javaid A, et al. Improving the multifunctionality of structural supercapacitors by interleaving graphene nanoplatelets between carbon fibers and solid polymer electrolyte. J Compos Mater. 2018;53(10):1401–1409.
[53] Wang B, et al. Graphene-based composites for electrochemical energy storage. Energy Storage Mater. 2020;24:22–51.
[54] Zhang R, Pang H. Application of graphene-metal/conductive polymer based composites in supercapacitors⋆. J Energy Storage. 2021;33:102037.
[55] Shao Y, et al. Graphene-based materials for flexible supercapacitors. Chem Soc Rev. 2015;44(11):3639–3665.
[56] Deka BK, et al. Multifunctional enhancement of woven carbon fiber/ZnO nanotube-based structural supercapacitor and polyester resin-domain solid-polymer electrolytes. Chem Eng J. 2017;325:672–680.
[57] Artigas-Arnaudas J, et al. Effect of electrode surface treatment on carbon fiber based structural supercapacitors: electrochemical analysis, mechanical performance and proof-of-concept. J Energy Storage. 2023;59:106599.
[58] Javaid A, et al. Carbon fibre-reinforced poly(ethylene glycol) diglycidyl ether based multifunctional structural supercapacitor composites for electrical energy storage applications. J Compos Mater. 2015;50(16):2155–2163.
[59] Javaid A, et al. Improving the multifunctional behaviour of structural supercapacitors by incorporating chemically activated carbon fibres and mesoporous silica particles as reinforcement. J Compos Mater. 2018;52(22):3085–3097.
[60] Reece R, Lekakou C, Smith PA. A structural supercapacitor based on activated carbon fabric and a solid electrolyte. Mater Sci Technol. 2019;35(3):368–375.

[61] Xu Y, et al. High-performance structural supercapacitors based on aligned discontinuous carbon fiber electrodes and solid polymer electrolytes. ACS Appl Mater Interfaces. 2021;13 (10):11774–11782.
[62] Deka BK, et al. Triboelectric nanogenerator-integrated structural supercapacitor with in situ MXene-dispersed N-doped Zn–Cu selenide nanostructured woven carbon fiber for energy harvesting and storage. Energy Storage Mater. 2021;43:402–410.
[63] Hubert O, Todorovic N, Bismarck A. Towards separator-free structural composite supercapacitors. Compos Sci Technol. 2022;217:109126.
[64] Dharmasiri B, et al. Flexible carbon fiber based structural supercapacitor composites with solvate ionic liquid-epoxy solid electrolyte. Chem Eng J. 2023;455:140778.
[65] Ma G, et al. High performance solid-state supercapacitor with PVA–KOH–K3[Fe(CN)6] gel polymer as electrolyte and separator. J Power Sources. 2014;256:281–287.
[66] Dong P, et al. A flexible solar cell/supercapacitor integrated energy device. Nano Energy. 2017;42:181–186.
[67] Wang Y, et al. Development of structural supercapacitors with epoxy based adhesive polymer electrolyte. J Energy Storage. 2019;26:100968.
[68] Li S, et al. Improved electrochemical and mechanical performance of epoxy-based electrolytes doped with mesoporous TiO2. Mater Chem Phys. 2018;205:23–28.
[69] González A, et al. Review on supercapacitors: technologies and materials. Renew Sust Energ Rev. 2016;58:1189–1206.
[70] Shirshova N, et al. Structural supercapacitor electrolytes based on bicontinuous ionic liquid–epoxy resin systems. ?J Mater Chem A. 2013;1(48):15300–15309.
[71] Shirshova N, et al. Composition as a means to control morphology and properties of epoxy based dual-phase structural electrolytes. J Phys Chem C. 2014;118(49):28377–28387.
[72] Pathak AK, et al. Relevance of graphene oxide as nanofiller for geometrical variation in unidirectional carbon fiber/epoxy composite. J Appl Polym Sci. 2021;138(38):50985.
[73] Manoj Kumar S, Kamal S. Effect of carbon nanofillers on the mechanical and interfacial properties of epoxy based nanocomposites: a review. Polymer Sci Series A. 2019;61(4):439–460.
[74] Huang F, et al. Creating ionic pathways in solid-state polymer electrolyte by using PVA-coated carbon nanofibers. Compos Sci Technol. 2021;207:108710.
[75] Carlson T, et al. Structural capacitor materials made from carbon fibre epoxy composites. Compos Sci Technol. 2010;70(7):1135–1140.
[76] Carlson T, Asp LE. Structural carbon fibre composite/PET capacitors – effects of dielectric separator thickness. Compos Part B Eng. 2013;49:16–21.
[77] Kim KM, et al. Supercapacitive properties of activated carbon electrode in potassium-polyacrylate hydrogel electrolytes. J Appl Electrochem. 2016;46(5):567–573.
[78] Sun J, et al. Mechanical and electrochemical performance of hybrid laminated structural composites with carbon fiber/ solid electrolyte supercapacitor interleaves. Compos Sci Technol. 2020;196:108234.
[79] Mapleback B, et al. Composite structural supercapacitors: high-performance carbon nanotube supercapacitors through ionic liquid localisation. Nanomaterials. 2022;12:2558.
[80] O'Brien DJ, Baechle DM, Wetzel ED. Design and performance of multifunctional structural composite capacitors. J Compos Mater. 2011;45(26):2797–2809.
[81] Zhou H, et al. A novel embedded all-solid-state composite structural supercapacitor based on activated carbon fiber electrode and carbon fiber reinforced polymer matrix. Chemical Engineering Journal. 2023;454:140222.