



## Review



## Sustainable electrochemical energy storage devices using natural bast fibres

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## ABSTRACT

Naturally abundant materials play a crucial role in the development of sustainable electrochemical energy storage (EES) devices including batteries and supercapacitors (SCs). This is due to limited available resources with regards to energy storage materials, and the environmental pollution produced by the toxic materials utilized in conventional EESs. In the current review, development in the field of natural bast fibres (jute, flax, hemp and kenaf) based EES devices performances is highlighted. This review emphasizes methods such as the direct use of modified fibres and activated carbon from biomass for the design of EES devices. Activated fibres were developed using both physical and chemical activation methods. Key challenges including active electrode materials preparation, capacitive retention, and the implementation of the fibre based EES devices are critically discussed. Furthermore, the recent surge in the use of wearables and portable technologies that demand further development of flexible/non-flexible EES devices are also explored. Future trends and perspectives on materials development, power management interface, recycling, biodegradability and circular economy are also addressed. It is concluded that the development of new renewable energy systems using bast fibres has many remarkable advances in device performance. For this, an innovative approach is required to develop high energy density bast fibre based sustainable EES devices which will be potentially implemented for clean energy solutions.

## 1. Introduction

The explosive growth in renewable energy sources and the global drive towards meeting the net-zero target have encouraged the development of long-life cycle sustainable electrochemical energy storage (EES) devices. To overcome the complexity, cost, and toxicity in the fabrication of electrodes in EES devices including batteries and supercapacitors (SC), new innovative ways of developing green energy storage devices are required. In this aspect, naturally abundant energy storage materials for the fabrication of flexible or non-flexible batteries and SCs offer many advantages to meet the requirements for the increasing demand in EES devices. Among the various electronic components development, the fabrication of EES devices such as batteries

and SCs using natural fibres have significant importance in sustainable electronic devices because they can overcome the issues related to the use of toxic materials in current batteries [1,2]. Focusing on green energy transition, the biocompatible materials based EES device fabrication can achieve the lowest possible environmental footprint of energy storage devices, as shown in Fig. 1a. Moreover, the recycling of biomass to active carbon electrodes for sustainable electronics products supports the circular economy research in advanced electronics technology [3,4]. Such EES devices could be further implemented for sustainable cities development. As per the UN sustainable development goals (SDGs), environmentally friendly EES research on energy materials has a significant impact on many SDGs as shown in Fig. 1b.

Maximum utilization of natural resources for the development of

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electronic devices can reduce hazardous and toxic electronic waste, which are a threat to the environment [5-10]. Recent technological innovations in green electronics have led to environmentally sustainable advances in new electronics technology [7,11-14]. For the design of green electronics, the development of natural fibres-based components has critical importance [15-20]. This is due to their intrinsic advantages such as low cost, excellent specific mechanical properties, natural abundance, and biodegradability [20-22]. Different types of electronic devices, including sensors, transistors, energy generators and EES, have been developed using natural fibres [2,23-28]. For this purpose, natural fibres have been engineered as dielectric, conductive, sensitive and active electrodes [29-34]. In cloth-based electronic device fabrication, natural fibre-based textiles have been directly used as a substrate [35-39]. In addition to this, other major implementations of natural fibres are the preparation of activated carbon from its biomass waste [40-45].

Compared to chemical vapor deposition and the Hummers method, biomass derived carbon methods are the simplest and most economically viable [46]. Through carbonization and activation using physical and chemical treatments, biomass becomes an excellent activated carbon material with the possibility of different chemical structures including carbon nanosheet (CNS) or honeycomb-like 3D carbon [47]. These activated carbon electrodes act as a free standing conductive network for growing metal oxides by using different synthesis methods, including hydrothermal preparation and electrodeposition [47]. The major advantage of conductive carbon networks with metal oxides is their contribution as high energy storage materials for the fabrication of EES, due to the influence of both the electrochemical double layer (EDL) formation and the pseudocapacitance of the material [47]. For the carbon fibre formation, bast fibres are widely available materials (areas of major production given in Table 1) among the natural fibres, and their rich fibrous nature offers advantages for electrode development and the formation of carbon fibre. For EES fabrication, high specific surface area (SSA) and porosity are highly important, providing large sites for formation of EDL [48-50]. In addition to this, the electrode conductivity, high tensile strength, and free-standing carbon structure without binders, give better performance for EES [51,52]. It was found that the bast fibre-based carbon exhibits all these properties and is therefore advantageous for EES development. For example, flax fibre based activated carbon exhibits an SSA of  $1649 \text{ m}^2 \cdot \text{g}^{-1}$  and pore size of 2 nm [53]. Due to these properties, both carbonized electrodes from biomass and coated (engineered) electrodes of bast fibres have been implemented for EES fabrication and reported widely. A detailed evaluation of the

**Table 1**

Major countries of production of bast fibres.

Type of fibre	Major countries of production	Ref
Jute	India and Bangladesh	[60,61]
Flax	Canada (oleaginous flax)	[61,62]
Hemp	Canada, China and the European Union (EU),	[61,63]
Kenaf	India and China	[64,65]
Ramie	China	[61,66]

fabrication and its performances as battery and SCs electrodes will pave the way for the development of new biocompatible EES. As compared to other reported reviews based on bast fibre composite preparation [54-57] and textile based EESs [58,59], in the present review we have focused on the major progress in implementations of bast fibres, especially jute, flax, hemp, and kenaf fibres as EES devices.

In this review paper, various methods employed for bast fibre based EES device fabrication and their performance are discussed. Following the introduction, in section 2 the importance of biomass-derived carbon and engineered bast fibres for EES fabrication are highlighted. The various fibres and adopted fabrication methods are discussed in section 3. In section 4, the future trends in this area are explored in detail and is followed by the concluding remarks.

## 2. Development of EESs from bast fibres through engineering of fibre and carbonisation

Bast fibres are derived from plant stem/bark/skin outer layers and some of the most used bast fibres include flax, jute, kenaf, hemp, and ramie. Bast fibres are the phloem or vascular tissues involved in the transport of carbohydrates, which grows along the stems of trees. The main three methods employed for bast fibre extraction from plants are (i) retting (ii) mechanical extraction and (iii) combined technique of retting and mechanical extraction. During this process, the outer layer (xylem and hurd/shives) is separated from other parts of the plant. A detailed study for the fibre extraction techniques is reported in another review article [61]. The chemical constituents of these fibres including cellulose, lignin and hemicellulose lead to excellent mechanical properties (tensile strength, elongation and Young's modulus range given in [67]) which is highly advantageous for the fabrication of various electronic components (such as sensors). The major constituents of these fibres are shown in Table 2 (data from [67]), including some physical

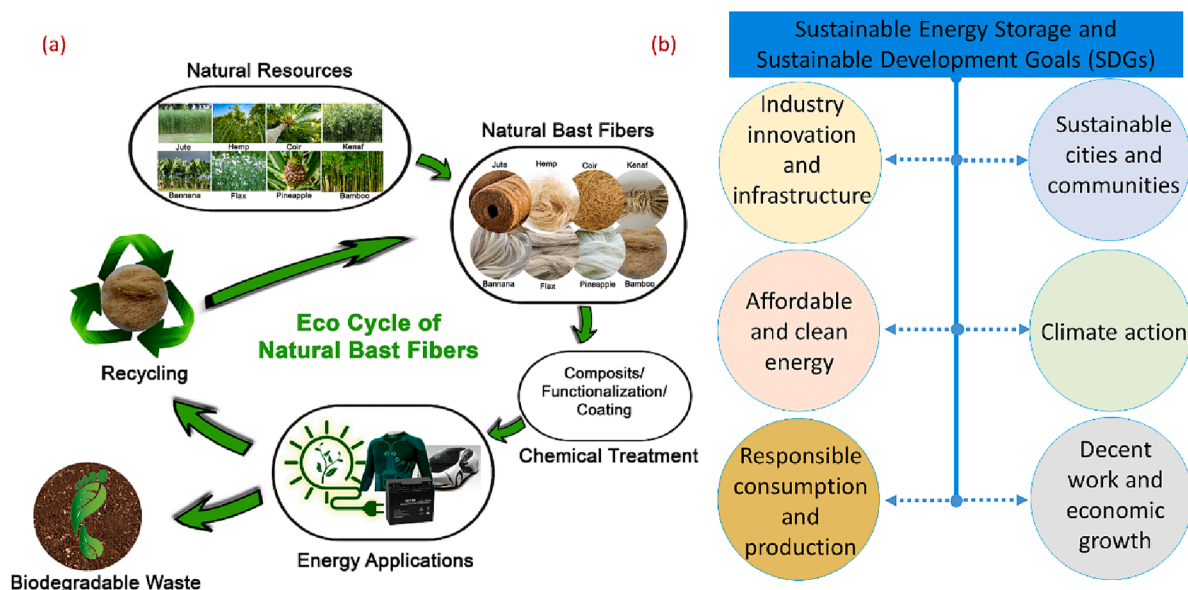


Fig. 1. (a) Sustainable energy storage system for a smart society (b) environmentally friendly energy storage and its scope in sustainable development goals (SDGs).

**Table 2**

The composition, physical and mechanical properties of jute, flax, hemp, and kenaf bast fibres [67].

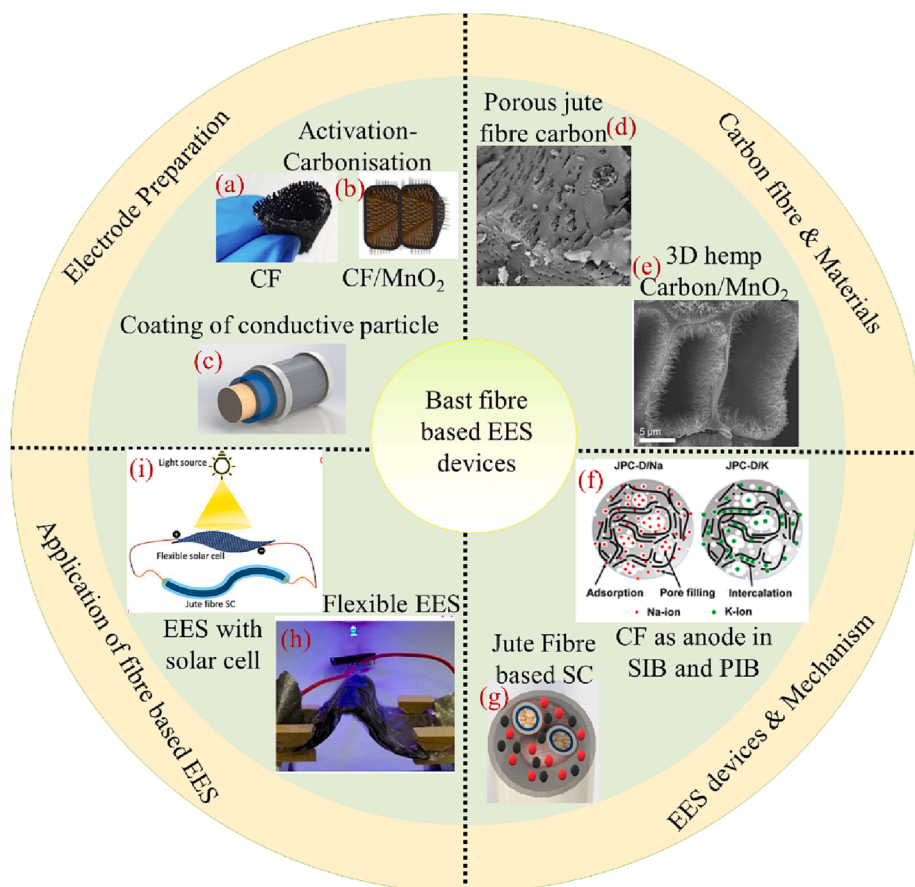
Fibres	Cellulose	Hemicellulose	Lignin	Diameter ( $\mu\text{m}$ )	Length (mm)	Tensile strength (MPa)	Young's Modulus (GPa)	Elongation at break (%)
Jute	64.4	12	11.8	5–25	0.8–6	400–800	10–30	1.8
Flax	64.1	16.7	2.0	5–38	10–65	800–1500	60–80	1.2–1.6
Hemp	74.4	17.9	3.7	10–51	5–55	550–900	70	1.6
Kenaf	53.4	33.9	21.2	12–36	1.4–11	295	–	2.7–6.9
Ramie	68.6	13.1	0.6	18–80	40–250	500	44	2

and mechanical properties as well.

Cellulose is the highest constituent in both, with its percentage higher in hemp and lower in kenaf fibres. Both, the mechanical properties, and the chemical percentage are highly influenced by the variety of the plant, the growth location, its age, the fibre source, as well as the fibre extraction method [61,68]. Thus, it should be noted that the displayed percentages may vary accordingly. The high cellulose content imparts efficient spinnability, tensile strength, stiffness, and stability, as well as softness, and hydrophilicity to the bast fibres. Additionally, the mechanical properties are mainly attributed to the cellulose content and its degree of polymerization, but also to the microfibril angle. Bast fibres with low microfibril angle and high cellulose amount have high tensile strength [54,61,67,69]. The mechanical properties of the fibres can be modified with graphene or graphene oxides for composite preparation [70]. Recently, these natural fibres were used as the active electrode material in EES devices (i) by synthesizing activated carbon from the biomass of natural fibres and (ii) by converting them into conductive electrode through coating materials such as polymers and carbons. Such electrodes were used for various flexible EESs development and implemented for various application and is reviewed in this manuscript, as shown in Fig. 2.

### 2.1. Activated carbon-based electrode from biomass of bast fibres

Any biomass consists of the three main components; cellulose, hemicellulose and lignin, and when calcined they may undergo different chemical routes at different temperatures. At a temperature of less than 100 °C, all the moisture evaporates out. Between 220 and 320 °C hemicellulose degrades, while cellulose degrades in the range of 320–400 °C. Lignin degrades at higher temperatures, with primary pyrolysis occurring at 200–400 °C and secondary pyrolysis at 400–600 °C [73]. After the removal of all the heteroatoms, the main residual is carbon [74]. Generation of carbon materials from biomass waste of natural fibres sources are very popular and usually advantageous over traditional chemicals as the sources are cheap, abundantly available, sustainable, and versatile. On the other hand, traditional production methods of activated carbon from non-renewable sources such as coal, peat, and petroleum are of high cost and harsh, despite giving a high yield [75]. The mechanical and electrical properties of the activated carbon and carbon composites from the biomass waste are quite attractive due to the organized structure of biomass in nature [76,77]. Biomass materials are widely used as electrode materials for SC applications because of the attractive carbon framework developed during



**Fig. 2.** The development and application stages of bast fibres based EES: (a-c) Electrodes preparation- (a and b) through carbonization and activation to form (a) carbon fibres, Reprinted from [49] with permission from Elsevier; (b) doping/reacting with pseudocapacitor material (e.g.  $\text{MnO}_2$ ) for high specific capacitance Reprinted (adapted) with permission from [47] Copyright (2017) American Chemical Society and (c) fibre/fabrics engineered to conductive electrodes by coating [24]. (d) morphology of jute derived porous carbon for anode of Li ion battery [71] Copyright (2019), with permission from Elsevier. (e) morphology of 3D structured carbon fibre form hemp and coated with  $\text{MnO}_2$  nanoparticle, Reprinted (adapted) with permission from [47] Copyright (2017) American Chemical Society. (f) charge storing mechanism in jute derived carbon as anode for sodium ion battery (SIB) and potassium ion battery (PIB), Copyright (2022) American Chemical Society [72]. (g) Fibre based SC with ionic distribution [24]. (h) application of fibre based flexible energy sources, Reprinted from Copyright (2020) [48] with permission from Elsevier. (i) fibre EES integrated with solar cell for energy system [24].



the conversion process, the complex pore structure and the greater SSA. For SC applications, hierarchical porous carbon with macropores combined with mesopores and micropores are advantageous because macropores offer minimal diffusive resistance during mass transport, while micropores and/or mesopores offer high SSA for active sites [78,79]. To achieve this carbon structure there are various methods by which biomass can be converted into activated carbon, and the choice of the method depends upon the application of the final product.

The traditional method of activating carbon consists of either physical or chemical activation. Physical activation is a two-step process in which carbonization takes place at less than 800 °C followed by activation at an elevated temperature (temperature range 900–1200 °C) in the presence of air, CO<sub>2</sub>, steam, etc [80]. On the other hand, in chemical activation, a precursor is mixed with an activating agent (KOH, NaOH, H<sub>2</sub>PO<sub>3</sub> etc) followed by calcination at a suitable temperature [81]. In both methods, activated carbon with a high SSA is obtained. However, the pores present are microporous in nature and are not suitable for the effective diffusion of electrolytes. In order to overcome this, hierarchically porous carbon (HPCs) (mixture of pores) are generated, which have gained recent attention as they are a mixture of macropores, micropores and mesopores. The macropore structure minimizes the diffusive resistance to mass transport and micro and mesopores offer a high SSA for active sites. To synthesize HPCs, one-pot methodology is used in which the carbon precursor is mixed with activating agent KOH, followed by a high temperature treatment (800–900 °C) [82]. The natural polymers in biomass contains cellulose, hemicellulose and lignin as well as some inorganic components (SiO<sub>2</sub>) that form a complex mixture, therefore it is challenging to develop a standard route to fabricate porous carbon materials. A simple activation route can result in microporous materials with well-defined shapes and structures. Hence, it is advantageous to add an activating agent which will create HPCs irrespective of the type of biomass used [82]. Such natural polymers are

insoluble in common solvents, therefore it would be useful to find a standard solvent and effective additives to develop a controlled synthesis of biomass-derived materials [83].

The surface area, pore size distribution, as well as the performance of the electrode material will be determined by the selected biomass precursors, the activation methods and the available functional groups on the surface [75]. In general, it is hard to define a perfect synthesis mechanism which suits to all kind of biomass precursors, hence while preparing any material, it is important to understand the application and the requirement. Thermal treatment is one of the most important techniques used for carbon preparation from biomass. For example, in one of the reported works, the physical, chemical and electrochemical properties of apple waste, peanut shell and corncob were measured in which all the materials were synthesized using an elevated temperature of 1100 °C for 1 hr [84]. After the preparation of materials, X-ray photoelectron spectroscopy (XPS) were performed to find out that the atomic surface chemical composition varies for all the samples due to different compositions of the biomass. Though the atomic surface oxygen for peanut and corncob was almost same (10.6, 10.3 wt%) but their type of oxygen functional groups were different. Moreover, peanut derived carbon has almost two times more carbonyl groups as compared to corncob which helps in increasing the surface wettability of carbon towards non-aqueous electrolyte [84]. It was observed that the performances of EES fabricated using bast biomass carbon depend on the type of activation and the composite used for enhancing the pseudocapitance and EDL [85,86]. As an example, different methods employed for carbonization of flax fibres, and incorporation with other materials including CNT and metal oxide for the development of the SC electrodes, are summarized in Fig. 3.


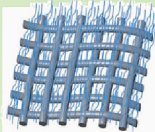



Activation methods for carbon fibre (CF) based electrode preparation	Prepared flax based active electrodes	Performances of EESs developed using flax fibre based electrodes
<b>CF/MnO<sub>2</sub></b> Direct carbonisation via temperature controlled reaction (maximum temperature 1000°C). MnO <sub>2</sub> nanosheet deposited via redox reaction between Carbon and KMnO <sub>4</sub>	(a) 	C <sub>sp</sub> : 683.73 F. g <sup>-1</sup> at 2 A. g <sup>-1</sup> ED: 46.54 W. h.kg <sup>-1</sup> CR : 94% after 1000 cycles
<b>CF/CNT</b> Carbonisation in tubular furnace 1000°C for 1 h with argon as a carrier gas	(b) 	C <sub>sp</sub> : 191 F. g <sup>-1</sup> at 0.1 A. g <sup>-1</sup> CR : 96% -5000 cycles-20 A. g <sup>-1</sup>
<b>CF, N-CF</b> Carbonisation in with nitrogen flow at 850°C for 2 h	(c) 	For CF [ C <sub>sp</sub> : 12.7 F. g <sup>-1</sup> For nitrogen doped CF [ C <sub>sp</sub> : 57.7 F. g <sup>-1</sup> at 2 mV. s <sup>-1</sup> ]
<b>CF-CO<sub>2</sub></b> Infrared-heating rapid thermal processing (700, 800°C) for 1 h in CO <sub>2</sub> at different heating rate	(d) 	C <sub>sp</sub> : 205 F. g <sup>-1</sup> (140 F. cm <sup>-3</sup> ) at 0.1 A. g <sup>-1</sup> (C <sub>sp</sub> ~3.4%-3000 cycles; CR: 97.7% -1000 bending cycles
<b>CF-NH<sub>3</sub></b> Carbonized at 800 °C-120 min, then activated NH <sub>3</sub> /N <sub>2</sub> in temperature (800, 850, 900°C) in different time duration	(e) 	C <sub>sp</sub> : 204 F. g <sup>-1</sup> at 0.5 A. g <sup>-1</sup> Energy density : 16.4 W.h. kg <sup>-1</sup> CR: 94% after 3000 cycles

Fig. 3. Summary of flax fibres based SCs development. (a) CF/MnO<sub>2</sub> Reprinted from [87] Copyright (2015), with permission from Elsevier. (b) CF/MNT Reprinted from [52] copyright (2017) John Wiley & Sons, Inc. (c) CF, nitrogen doped CF Reprinted from [51] Copyright (2017), with permission from Elsevier. (d) CF with CO<sub>2</sub> activation, Reprinted from [49] with permission from Elsevier. (e) CF with NH<sub>3</sub> activation, Reprinted from Copyright (2020) [48] with permission from Elsevier.



## 2.2. Conductive bast fibre electrode prepared by coating with materials

It has been observed that for bast fibre based carbonized electrode fabrication, high temperature processing and physical/chemical activation are required when using the biomass waste. The need for high temperature processes can be avoided by converting the insulative fibre into conductive fibre by functionalizing with conductive materials. There is limited work reported for such electrode and its EES fabrication. To make a conductive electrode, in one reported method, the jute fibre was treated with Ag nanoparticle (jute fibre dipped in  $\text{AgNO}_3$  solution and treated with UV light), thus increasing the conductivity [88]. It was found that the Ag nanoparticle functionalization leads to a reduction of resistance by 15,000 times as compared to that of bare jute fibre. Such enhanced conductive jute fibre shows strong opportunities for application in smart conductive textiles [88]. In another work, conductive polymer (PEDOT:PSS) and SWCNTs were coated by dip coating for SC development (discussed in sect. 3.1).

This section shows that there are two main methods of electrode development from bast fibres including carbon using biomass and coated electrodes using conductive materials. These electrodes show excellent electrical and structural properties including high specific surface area and porosity. The biomass derived activated carbon has several industrial applications ranging from wastewater treatment to  $\text{CO}_2$  adsorption, energy storage applications etc [89,90]. The advantageous bast fibre electrodes for EES fabrication are discussed in the

following section.

## 3. Bast fibres for EES development

In this section, we have discussed various bast fibres utilised for the EES design. Among the various works on bast fibre based EES devices, we noted that the key development was reported in SCs and works related to batteries or Li-ion based storage are limited, revealing requirements for developing new devices in this area. Some of the important reports of bast fibre with Li-ion based storage include battery and capacitors, developed using porous jute fibre carbon for Li ion battery [71], sodium and potassium ion battery [72] and carbon derived from kenaf fibre for Li-ion capacitors [91]. In EES development, the specific surface area, pore size and distribution, interconnected network of carbon layers, conductivity and surface roughness of the electrode influence the electrochemical performance of the EES devices. A comprehensive details of various methods for fabricating EES devices is presented, providing a comparative study.

### 3.1. Jute fibre based EES

Jute is the second most produced natural fibre with typically 70 wt% of cellulose, 15 wt% of hemicellulose and 10 wt% of lignin [24,92]. Both jute derived carbon and jute fibre based conductive electrodes have been used for EES fabrication especially for the development of SCs. By using

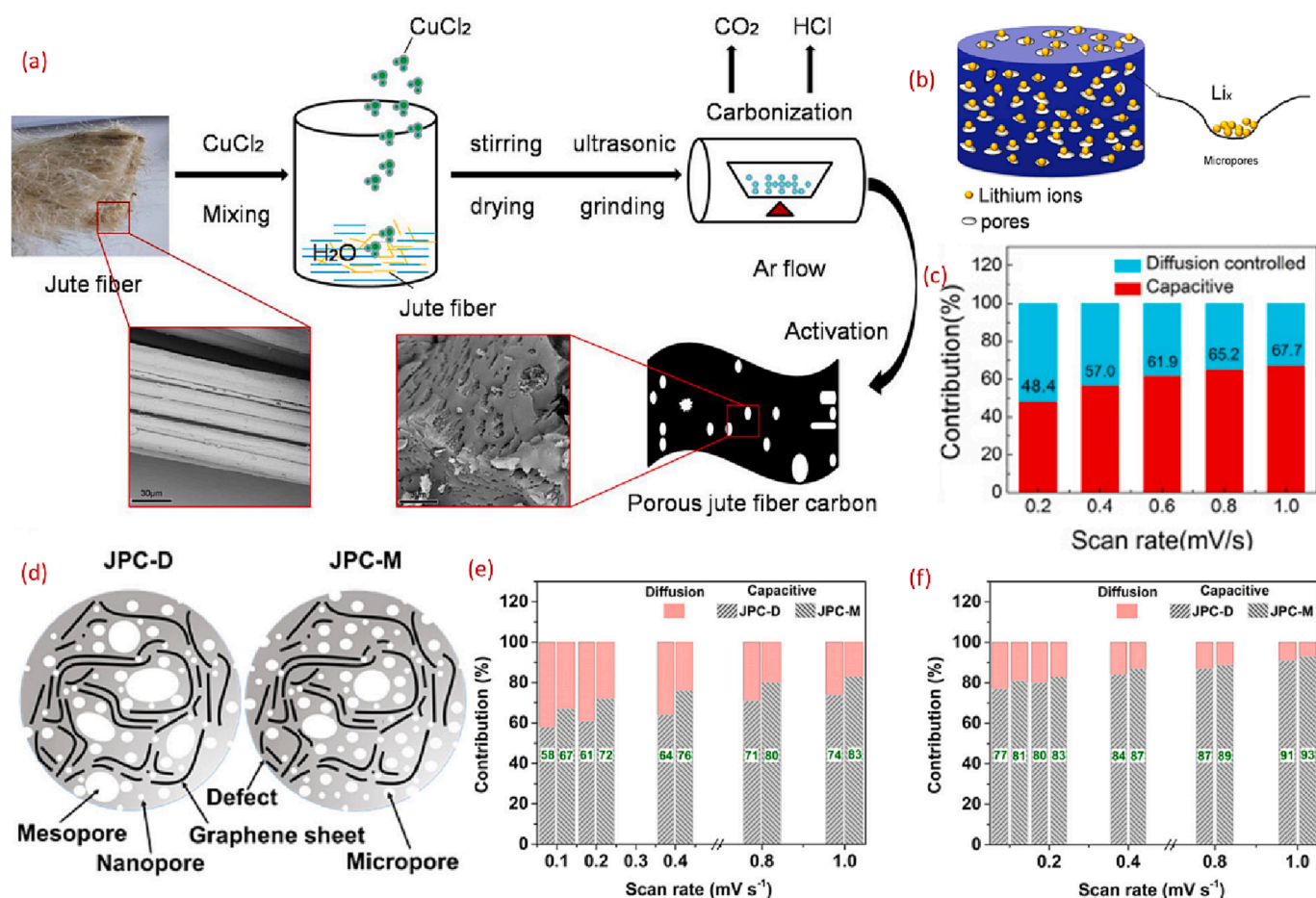


Fig. 4. Procedure for the preparation of jute derived carbon for the development of anode of Li ion battery, Reprinted from Publication [71] Copyright (2019), with permission from Elsevier. (b) schematic representation of Li ion diffusion in mesopores of the electrode, Reprinted from Publication [71] Copyright (2019), with permission from Elsevier. (c) Capacitive and diffusion controlled behaviors contribution for capacitance of the jute based Li ion battery at various scan rates Reprinted from Publication [71] Copyright (2019), with permission from Elsevier. (d) Schematic represents the pore structure of JPC-D and JPC-M [72] (e) diffusive and capacitive contribution of charge storage of JPC-D and JPC-M in sodium system [72] (f) diffusive and capacitive contribution of charge storage of JPC-D and JPC-M in potassium system [72]. (d-e) Copyright (2022) American Chemical Society [72].

different approaches, jute derived carbon has been prepared and is summarized recently [93]. It was noted that the intrinsic fibrous structure of jute is beneficial for the fabrication of energy storage devices due to the ionic conductivity and jute derived carbon has been employed as an electrode in battery and SCs [71,94]. For Li battery development, graphitic porous carbon sheets (GPCS) were synthesized from jute stick biomass [95]. It was found that the graphitic sheet morphology enables to store more Li ions and the electrolyte ions diffuse very easily, resulting in the enhancement of the electrochemical performances [95]. The GPCS based anode on Li battery exhibits a specific capacity of 1022 mA h g<sup>-1</sup> after 10 cycles at 100 mA. g<sup>-1</sup>. The device also retains a high capacity (541 mA. h.g<sup>-1</sup>) after 250 cycles at a high current of 500 mA. g<sup>-1</sup>[95]. For an anode electrode in a Li ion battery, jute derived carbon was prepared by activation with CuCl<sub>2</sub> which offered high porosity and high SSA (2043.528 m<sup>2</sup>. g<sup>-1</sup>). The fabrication procedure is shown in Fig. 4a [71]. The porosity enhances the pathways for Li<sup>+</sup> ions and large surface area increases the electrode/electrolyte contact area. Fig. 4b shows the macropores due to etching with CuCl<sub>2</sub> for supporting Li<sup>+</sup> ions diffusion [71]. Due to the excellent property of the anode materials, the battery exhibited a specific capacity of 580.4 mA. h. g<sup>-1</sup> at a current density of 0.2C [71]. The capacitive and diffusive controlled behavior that were studied in different scan rates, showed an enhanced capacitance of the device as depicted in Fig. 4c [71]. In another recent work, a porous hard carbon (HC) derived from jute were used as an anode for sodium ion battery (SIB) and potassium ion battery (PIB)[72]. Authors prepared two types of jute derived HC (JPC): one via direct carbonization, known as JPC-D, and another one microwave pre-treated, known as JPC-M. The resulted JPC-D shows micro-mesoporous structure and the JPC-M exhibits ultra-microporous structure, as shown in Fig. 4d [72]. It was found that the presence of more mesoporous structure leads to achieve high reversible capacity with excellent rate performances and life cycle for SIB and PIB. The HC prepared from the jute shows that for SIB the main mechanism leading to charge storage are (i) adsorption (ii) intercalation and (iii) pore filling. However, for PIB, the main mechanism for charge storing is (i) adsorption and (ii) intercalation. Furthermore, it was found that the non-Faradaic process is pivotal for both JPC-D and JPC-M in case of SIB and PIB as well [72]. The capacitive or diffusive contribution is varied with mesoporous structure of the HC. Fig. 4e shows the comparison of JPC-D and JPC-M in sodium system. The capacitive contribution for JPC-M (67%) is higher than that for JPC-D (58%). A similar trend was observed in potassium system as shown in Fig. 4f. Moreover, the capacitive contribution is higher for potassium

system compared with the sodium one, in both JPC-D and JPC-M. The fabricated SIB shows a specific capacity of 328 mA. h. g<sup>-1</sup> and is higher compared to that of the PIB (245 mA. h. g<sup>-1</sup>) [72].

Several studies have been carried out for the development of jute derived carbon-based SCs, and several of them are given in Table 3. In most of the works, KOH has been used as an auxiliary agent for the preparation of the carbon from jute [40,94,96-98]. In one report, it was found that as compared to other activated carbon from sugar and rice husk, the activated carbon from jute exhibits a high capacitance (476F. g<sup>-1</sup> at a current density of 0.2 A. g<sup>-1</sup>). Not only the activation process results in a larger specific surface area for jute, but the high capacitance is also allocated to the presence of quinone-hydroquinone species that contribute to the pseudocapacitance [97]. For SC fabrication, the jute-derived carbon has been prepared by a facile hydrothermal method (180 °C) and chemical activation using KOH at 800 °C under an argon atmosphere [99]. The prepared, carbonized jute powder (as shown in Fig. 5a) was converted into a paste and applied on the top of a nickel electrode for SC fabrication [99]. Such carbonized jute fibre shows a porous structure due to the activation with KOH and it also shows the presence of a graphite conducting phase, making the carbonized jute an excellent electrode for SC. Electrochemical analysis (cyclic voltammogram given in Fig. 5b) shows that in a three-electrode system measurement, the carbonized jute fibre exhibits a specific capacitance of 408F. g<sup>-1</sup> in a KOH electrolyte, as shown in Fig. 5c [99]. The full SC device, comprised of two carbonized jute electrodes, shows a specific capacitance of 51F.g<sup>-1</sup>, as shown in Fig. 5d. It was noted that the energy storing performance depends on the measurement temperature (as presented in Fig. 5e), with the capacitance changing with temperature. Such carbonized jute fibre-based SC electrodes exhibited 100% retention in charge storage capacity after 5000 charging/discharging cycles and were not heavily influenced by different bending angles [99].

In addition, with derived jute fibre-based electrodes, modified jute fibre with conductive polymers was also used for the development of SCs. In previous work, jute fibre was functionalized with conductive polymer PEDOT: PSS for SC fabrication [24]. The high cellulose content in the jute has the advantage of absorbing a high quantity of the aqueous active electrode ink into the surface and bulk of the fibre. The SCs were fabricated by coating PEDOT: PSS on the top of this jute fibre and to further improve the performance, SWCNTs were coated on the top of the PEDOT: PSS. In the sustainable development of EES, not only the active electrode materials and substrate, but also the electrolyte should be biocompatible/sustainable. In this aspect, this work showed the use of a

**Table 3**  
Comparison of performances of bast fibre based EESs.

Fibres	Electrode	SSA	Electrolyte	Specific Capacitance	Energy Density	Power Density	Life Cycle	Ref
Jute	Activated Carbon-Jute	825 m <sup>2</sup> .g <sup>-1</sup>	0.5 M H <sub>2</sub> SO <sub>4</sub>	476F. g <sup>-1</sup>			350	[97]
Jute	Carbonized jute	1769 m <sup>2</sup> . g <sup>-1</sup>	3 M KOH	185F. g <sup>-1</sup> (500 mA. g <sup>-1</sup> )	21 W. h. kg <sup>-1</sup>	1.82 kW. kg <sup>-1</sup>	5000	[99]
Jute	PEDOT:PSS/ SWCNT coating		HEC-KCl	8.65 mF. cm <sup>-1</sup>	0.712 μW. h. cm <sup>-1</sup>	3.85 μW. cm <sup>-1</sup>	5000	[24]
Flax	PNCF	1152 m <sup>2</sup> . g <sup>-1</sup>	6 M KOH	204.1F. g <sup>-1</sup>	16.4 W. h. kg <sup>-1</sup>	100 W. kg <sup>-1</sup>	3000	[48]
Flax	MnO <sub>2</sub>	25.6 – 33.6 m <sup>2</sup> . g <sup>-1</sup>	0.1 M Na <sub>2</sub> SO <sub>4</sub>	683.73F. g <sup>-1</sup>	46.54 W. h. kg <sup>-1</sup>	45.50 kW. kg <sup>-1</sup>	10,000	[87]
Flax	Nitrogen doped flax fibre	410 m <sup>2</sup> . g <sup>-1</sup>	1 M H <sub>2</sub> SO <sub>4</sub>	57.7F. g <sup>-1</sup>				[51]
Flax	CNT coated Flax	580 m <sup>2</sup> . g <sup>-1</sup>	6 M KOH	191F. g <sup>-1</sup>		3.3 kW. kg <sup>-1</sup>	5000	[52]
Flax	Flax derived AC	1649 m <sup>2</sup> .g <sup>-1</sup>	H <sub>2</sub> SO <sub>4</sub>	189F. g <sup>-1</sup>			150 k	[101]
Flax	AC	707 m <sup>2</sup> . g <sup>-1</sup>	6 M KOH	205F. g <sup>-1</sup>			3000	[49]
Hemp	CO <sub>2</sub> activated HFPC	1060 m <sup>2</sup> . g <sup>-1</sup>	PVA-KOH	457F. g <sup>-1</sup>	25.3 W. h. kg <sup>-1</sup>	4320 W. kg <sup>-1</sup>	10,000	[50]
Hemp	Carbon Nanosheet	1505 m <sup>2</sup> . g <sup>-1</sup>	Ionic liquid	113F. g <sup>-1</sup> –20 °C 144F. g <sup>-1</sup> –60 °C 142F. g <sup>-1</sup> –100 °C	19 W. h. kg <sup>-1</sup> –20 °C 34 W. h. kg <sup>-1</sup> –60 °C 40 W. h. kg <sup>-1</sup> –100 °C	28 kW. kg <sup>-1</sup> –20 °C 49 kW. kg <sup>-1</sup> –60 °C 77 kW. kg <sup>-1</sup> –100 °C	10,000	[46]
Hemp	MnO <sub>2</sub> nanowire and HC	438 m <sup>2</sup> . g <sup>-1</sup>	1 M Na <sub>2</sub> SO <sub>4</sub>	340F. g <sup>-1</sup>	33.3 W. h. kg <sup>-1</sup>	14.8 kW. kg <sup>-1</sup>	3000	[47]
Kenaf	Kenaf steam derived carbon	1480 m <sup>2</sup> . g <sup>-1</sup>	0.1 M H <sub>2</sub> SO <sub>4</sub>	327F. g <sup>-1</sup>	16 W. h. kg <sup>-1</sup>	200 W. kg <sup>-1</sup>	5000	[119]
Kenaf	MnO <sub>2</sub> /3D-PC		1 M Na <sub>2</sub> SO <sub>4</sub>	416F. g <sup>-1</sup>	17.3 W. h. kg <sup>-1</sup>	198 W. kg <sup>-1</sup>	1000	[110]
Kenaf	3D-KSC/rGO/PANI		0.5 M H <sub>2</sub> SO <sub>4</sub>	1224F. g <sup>-1</sup>	144.4 W. h. kg <sup>-1</sup>	0.218 W. kg <sup>-1</sup>	5000	[112]
Kenaf	PNI/KPC		0.1 M H <sub>2</sub> SO <sub>4</sub>	113F. g <sup>-1</sup>			2000	[111]

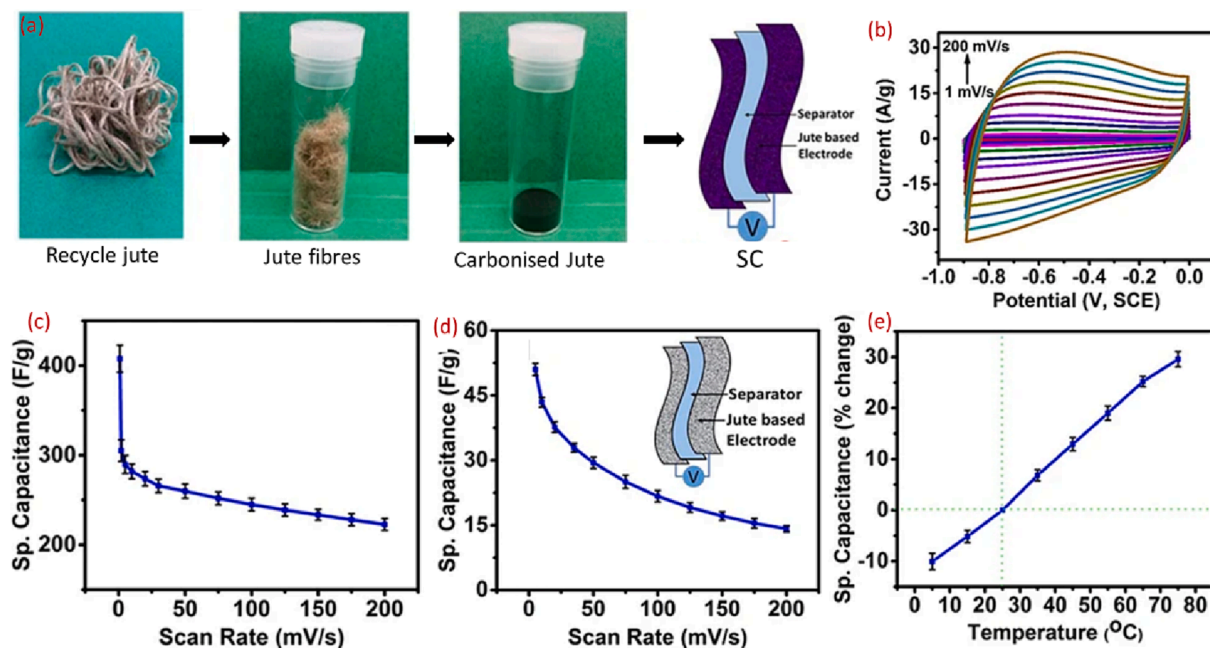


Fig. 5. (a) Schematic of the carbonized jute-based SC fabrication process [99] (b) CV curves and its (c) variation of specific capacitance for carbonized jute in a three-electrode measurement system [99]. (d) and (e) specific capacitance and capacitance changes with temperature for a carbonized jute based SC device [99].

new electrolyte based on hydroxyethyl cellulose-potassium chloride (HEC-KC) gel. This gel was sandwiched between two identical electrodes with a cellulose-based separator. The schematic of the SC is shown in Fig. 6a [24]. The electrochemical studies predict that during charging, the ions are distributed on the electrode surface leading to a high capacitance ( $17 \text{ mF.cm}^{-1}$  at  $1 \text{ mV.s}^{-1}$ ) as shown in Fig. 6b and 6c. The studies reveal that the pseudocapacitance due to a redox reaction of the conjugated polymer and EDL formation contribute to a capacitance enhancement of the device [24].

### 3.2. Flax fibre based EES

Flax, one of the most abundant fibres in the world, widely used in textiles, biomedical applications, and automotive cushioning, was recently found to be an excellent biomass material for EES device fabrication [100,101]. Compared with other fibres, flax is relatively stronger in terms of mechanical strength, offers good absorbent properties, making them a faster drying Fibre than cotton that may be used as a free-standing carbonized substrate for electrode development [52,100]. Various methods employed for the activation of flax fibre for carbon-based electrodes in SC developments are shown schematically in Fig. 3. The SC realized from carbon fibre cloth derived from flax fibres exhibits a specific capacitance of  $0.78 \text{ F.g}^{-1}$  at  $0.1 \text{ A.g}^{-1}$  and 100%

capacitance retention after  $10^4$  cycles charging/discharging at  $5 \text{ A.g}^{-1}$  [87]. This excellent stability and electrochemical performances led to the development of flexible SCs with high energy density and capacitance by modification with pseudocapacitor materials. As an example,  $\text{MnO}_2$  deposited on top of the flax fibre based carbon cloth (schematic shown in Fig. 7a) was reported as the first SC electrode on this flax textile derived carbon fibre, with a specific capacitance of  $683.73 \text{ F.g}^{-1}$  at  $2 \text{ A.g}^{-1}$ , (the capacitance variation with current density is shown in Fig. 7b) and a high energy density of  $46.54 \text{ W.h.kg}^{-1}$  [87]. The SC also shows capacitance retention ratio above 94% after 1000 cycles and the results suggest its potential application in wearable energy sources [87]. In another work, the authors deposited CNTs on a carbonized flax fabric (fabrication step shown in Fig. 7c), offering the advantage of high conductivity and chemical stability when compared to the flax fibre-metal oxide hybrid electrodes [52]. The CNTs provide micropores and mesopores for ion transport on the fabric. In addition to this, *in situ* growth of CNTs offers binder-less coating of the active electrode, which in turn reduces the ohmic resistance and enhances the cyclic rate capability. Due to the high conductive pathways for fast electron transport of carbon fibres and CNTs, excellent SC performance can be obtained with a specific capacitance of  $191 \text{ F.g}^{-1}$  at  $0.1 \text{ A.g}^{-1}$ . As compared to 1000 cycles of carbonized flax fibre-  $\text{MnO}_2$  [87], this CNT based flax Fibre carbonized SC exhibits a high cycling retention of 96%

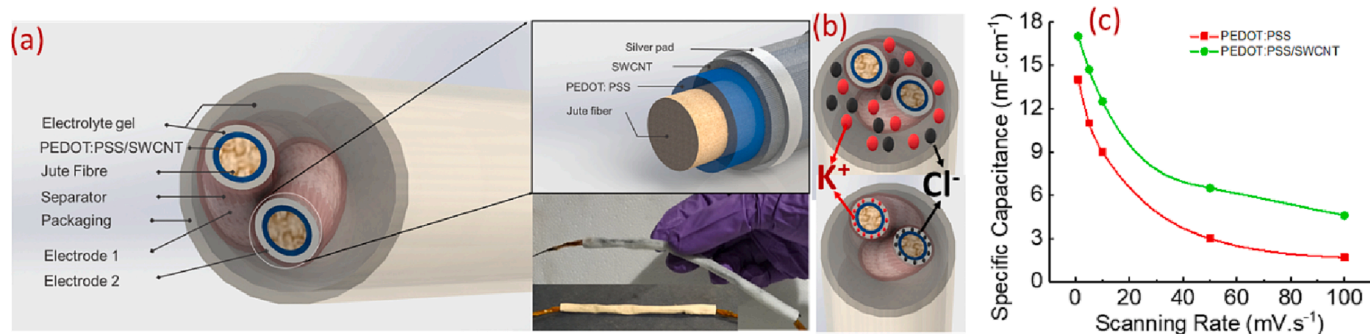
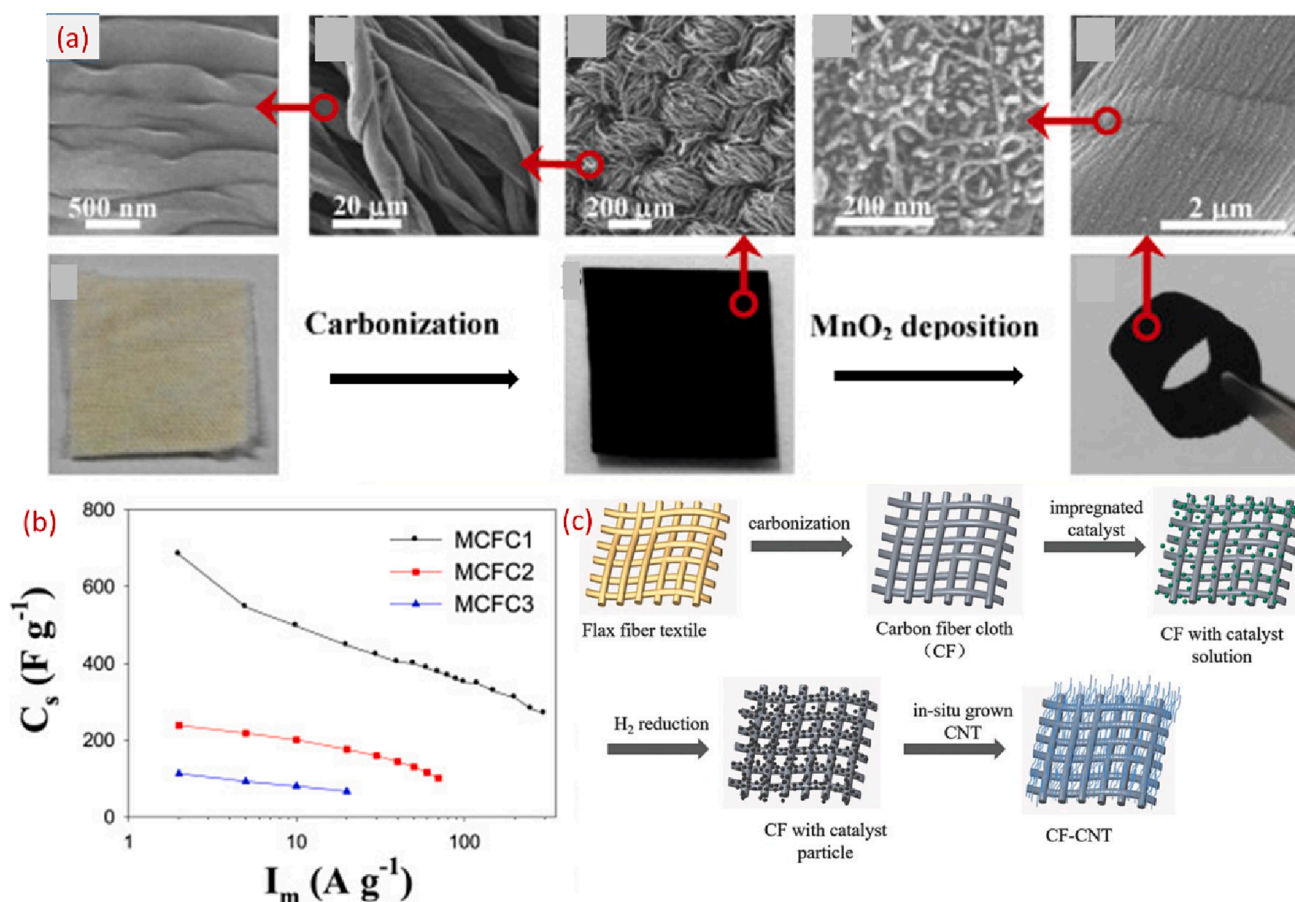


Fig. 6. (a) Schematic of PEDOT:PSS/ SWCNT coated jute fibre based SC [24] (b) ionic distribution in the jute fibre SC electrodes [24] (c) comparison of specific capacitance of variation of PEDOT:PSS/SWCNT and PEDOT:PSS based Jute SCs [24].





**Fig. 7.** Preparation method of flax Fibre based carbon electrode (a) Schematic illustration of the preparation process of MnO<sub>2</sub>/carbon fibre cloth (MCFC) hybrids from flax fabric and the SEM images of the prepared cloth, Reprinted from [87] Copyright (2015), with permission from Elsevier. (b) Specific capacitance plots of the (MCFC1, MSFC2, MCFC3 represent the concentration of KMnO<sub>4</sub> (aq) from 1, 2–5 mM) composites at different current densities, Reprinted from [87] Copyright (2015), with permission from Elsevier. (c) Flax fibre based electrode prepared for the SC development for that carbon fibre – carbon nanotube (CF-CNT) hybrids were prepared, Reprinted from [52] copyright (2017) John Wiley & Sons, Inc.

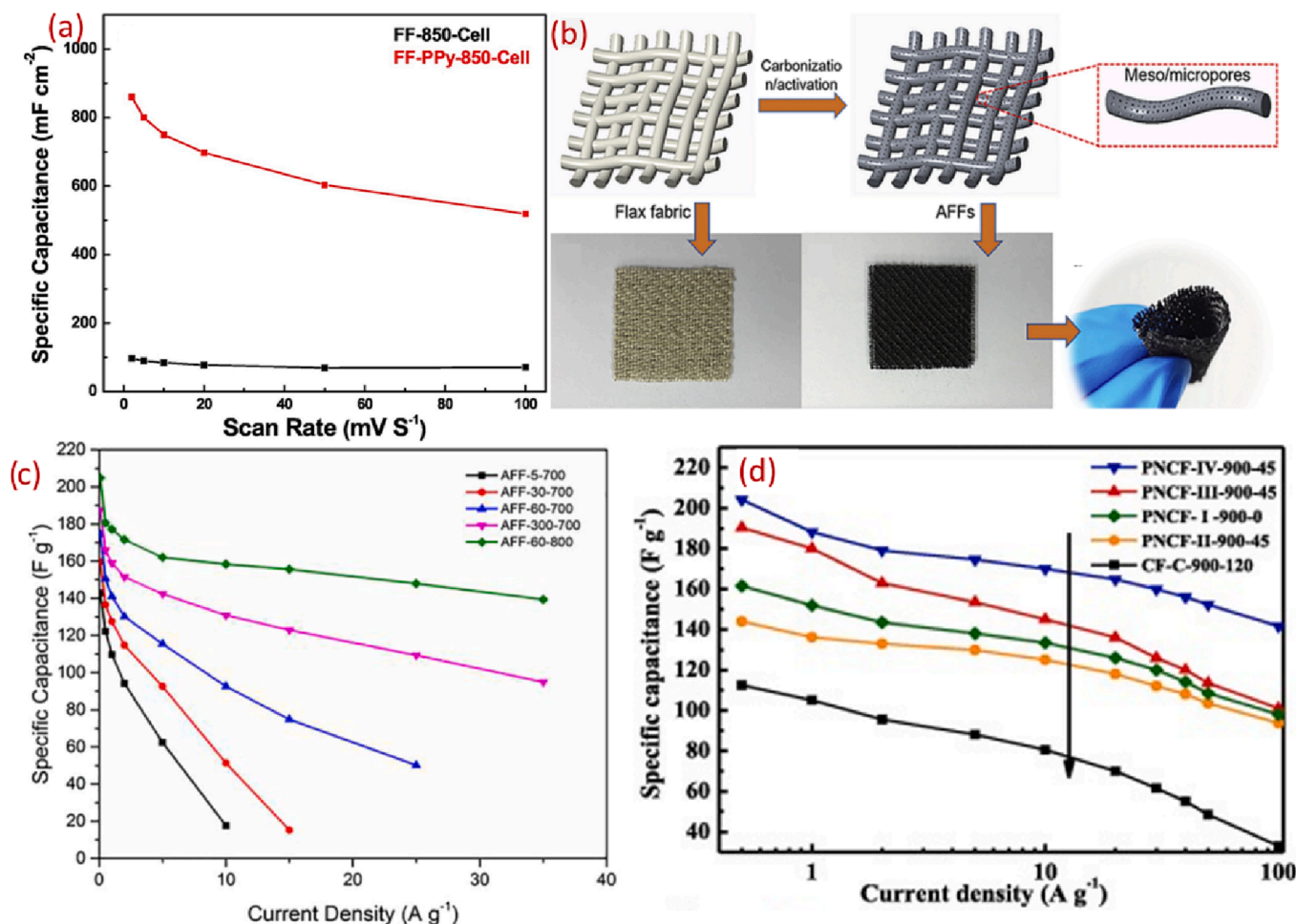
after 5000 cycles [52]. Along with coating or *in situ* growth of metal oxide and CNTs, the porous structure and large specific surface area of the carbonized Fibre influence increasing performance of the specific capacitance of the SC. In addition to this, it was found that heteroatom doping (nitrogen doping) may also lead to an increase in the capacitance value due to the presence of both mesopores and micropores of the nitrogen doped carbonized materials, as compared to the undoped carbon electrode [51].

In a recent study, flax fibres were converted into carbonized fibres by thermal treatment (550, 700, 850, and 1000 °C) in a nitrogen atmosphere [51]. It was noted that the processing temperature influences the carbon conversion, and at 850 °C the carbon content reaches ~ 91.8 wt %. Furthermore, this carbonized electrode was treated with pyrrole to obtain nitrogen doped carbon Fibre from flax. The electrochemical comparison studies show that, due to the high porosity and pseudocapacitance influences, nitrogen doped carbon fibres exhibit a high capacitance of 860 mF. cm<sup>-2</sup> (57.7F. g<sup>-1</sup>) at 2 mV. s<sup>-1</sup> as compared to 97 mF. cm<sup>-2</sup> (12.7F. g<sup>-1</sup>) for carbon fibre based SC, as shown in Fig. 8a [51]. Nitrogen doping is shown to increase the electrical conductivity of carbon fibres for enhancing the electrochemical performances [48].

In carbon based SCs, the specific surface area with high meso/micropore ratios are advantageous for charge accumulation and enhancing the energy density. In a recent work (Fig. 8b) it was reported that the heating rate and direct carbonization/activation in a CO<sub>2</sub> atmosphere can control the surface area and meso/micropore ratios of carbonized fibre [49]. The heating rate influences the mesopore volume/total pore volume ratio allowing the optimization of the

electrochemical performance. For a high heating rate such as 300 °C/min at a temperature of 700 °C, the activated flax fabric exhibits (AFF-300–700) a CV curve of quasi-rectangular profile (typical EDL formation) compared with a triangular shape at 5 °C/min [49]. This enhancement in the performance leads to an increase in the specific capacitance of the devices as shown in Fig. 8c. The authors found that the activated flax fabric heated at 60 °C/min for 800 °C (AFF-60–800) exhibited 61% of mesoporosity, with 707 m<sup>2</sup>. g<sup>-1</sup> SSA and showed capacitance of 205F. g<sup>-1</sup> (140F. cm<sup>-3</sup>) at 0.1 A. g<sup>-1</sup>. Furthermore, 97.7% of SC retention after 1000 bending cycles, and ~ 3.4% capacitance loss after 3000 charging/discharging cycles at 10 A. g<sup>-1</sup> were reported. This heating rate study under CO<sub>2</sub> activations has led to further investigation of other fibres in different heating and activation conditions [49]. It was observed that at certain temperature through CO<sub>2</sub> activation and nitrogen doping, the surface area, mesoporosity and electrical conductivity of the activated flax fibre could be modified and controlled.

To overcome the complex, time consuming and costly synthesis route of nitrogen-containing precursors, the treatment of flax in ammonia (which act as both activating agent and nitrogen dopant reacting) was explored, allowing porous nitrogen-doped flexible carbon Fibre sheets (PNCFs) to be developed for SC fabrication [48]. The changing electron-donor properties and electrode surface chemical properties enhance the electrochemical performances of EES devices [48,102]. It has been found that the incorporation of heteroatoms into carbon is one of the best methods [48]. Heteroatom (nitrogen) doping in carbon reduces the charge transfer resistance and increases wettability [48]. This leads to



**Fig. 8.** Performance of flax Fibre based SCs (a) Specific capacitance variation calculated on the basis of the cyclic voltammetry data at different scan rates for flax fabric (FF) based SC using FF carbon and FF treated with polypyrrole (PPy) developed at 850 °C, Reprinted from [51] Copyright (2017), with permission from Elsevier. (e) Preparation process of the activated flax fabric (AFF) electrode with controlled meso/microporous structure and the digital photographs Reprinted from [49] with permission from Elsevier. (f) Specific capacitance variation at different current densities for AFF processed at different temperatures, Reprinted from [49] with permission from Elsevier. (g) Specific capacitance variation at a current density of 1  $\text{A g}^{-1}$  of nitrogen doped flexible carbon Fibre (PNCF) from flax in different activation conditions, Reprinted from Copyright (2020) [48] with permission from Elsevier.

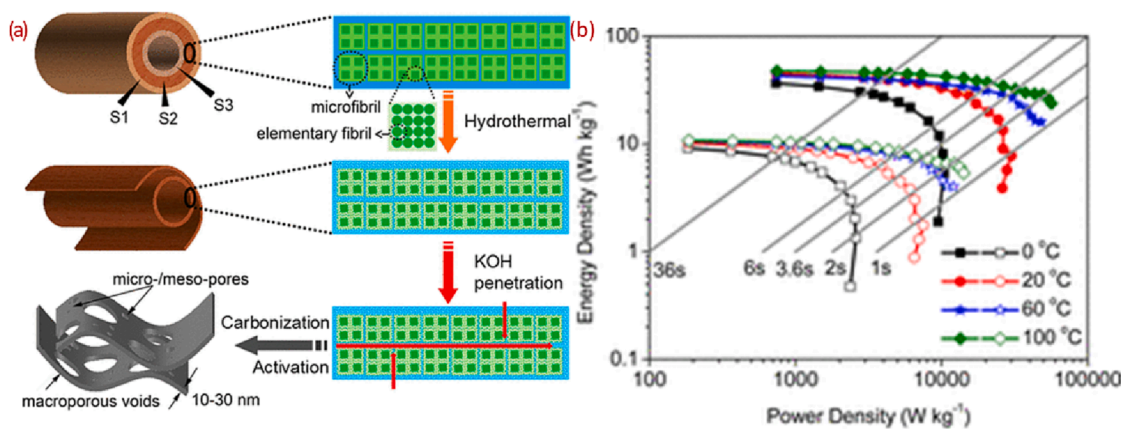
an enhancement in the capacitance of associated EES devices. Ammonia activation and nitrogen doping applied to flax fibre based SCs result in a specific capacitance of 204.1  $\text{F g}^{-1}$  at a current density of 0.5  $\text{A g}^{-1}$  and energy density of 16.4  $\text{W h kg}^{-1}$  [48]. The activation temperature, duration time and conditions for ammonia switching in activation, influenced the energy storage performance as shown in Fig. 8d [48]. Flax derived carbon based SCs have demonstrated comparatively low life cycle (majority of them reported 5000 charging/discharging cycles), as compared to activated carbon or graphene based SCs. These issues were overcome through the introduction of new preparation methods such as combined (1) hydrothermal conversion and (2) pyrolytic-chemical activation by KOH, in which the developed SC showed 150,000 charging/discharging cycles with 85% of capacitive retention at a current density of 5  $\text{A g}^{-1}$ . The synthesized carbon devices exhibited specific capacitance of 500  $\text{F g}^{-1}$  (current density of 0.25  $\text{A g}^{-1}$ ) and 189  $\text{F g}^{-1}$  (current density of 0.5  $\text{A g}^{-1}$ ), respectively, with 3 electrode and 2 electrode system measurements [101].

### 3.3. Hemp fibre based EES

Hemp, which is grown without any special requirements such as climate, pesticides, or fertilizers, have been widely used for building materials, paper, plastics, food, medicine etc. Physical and chemical activation methods have been utilized for the preparation of activated

carbon fibres from hemp and it produces excellent porous electrodes for SC fabrication [103]. Synthesis included physical activation with steam, and chemical activation with  $\text{ZnCl}_2$  or  $\text{H}_3\text{PO}_4$  [104–106]. To achieve interconnected two-dimensional graphene-like carbon nanosheets with mesoporosity for enhanced electrochemical performance of the SCs, a hydrothermal process combined with activation with KOH was used in hemp fibre carbonized electrode formation, as presented in Fig. 9a. During the hydrothermal process, the outer and inner layer which mostly consist of lignin (S1) and semi-cellulose (S3) respectively, become hydrolyzed and the middle layer, which consist of crystalline cellulose (S2), becomes partially carbonized, as depicted in Fig. 9a [46]. The KOH treatment during chemical activation (700–800 °C) followed with hydrothermal processing results in layer separation and forms micro/mesoporosity. SCs based on such carbon CNS in ionic liquid electrolytes reveal excellent energy storage performance as compared to activated graphene or activated carbon based SCs. The full SC device based on CNS electrodes prepared at 800 °C showed a maximum energy density of 12  $\text{W h kg}^{-1}$ , which is highlighted as being higher than commercial SCs, and these could be charged in 6 s. Fig. 9b shows the comparison of energy and power densities for the total mass of device (hollow, full device) and the mass of the active material (solid, single electrode) at various temperatures [46].

In EES device fabrication, 3D nanostructured materials show excellent electrochemically active surface area for EDL formation. Recent

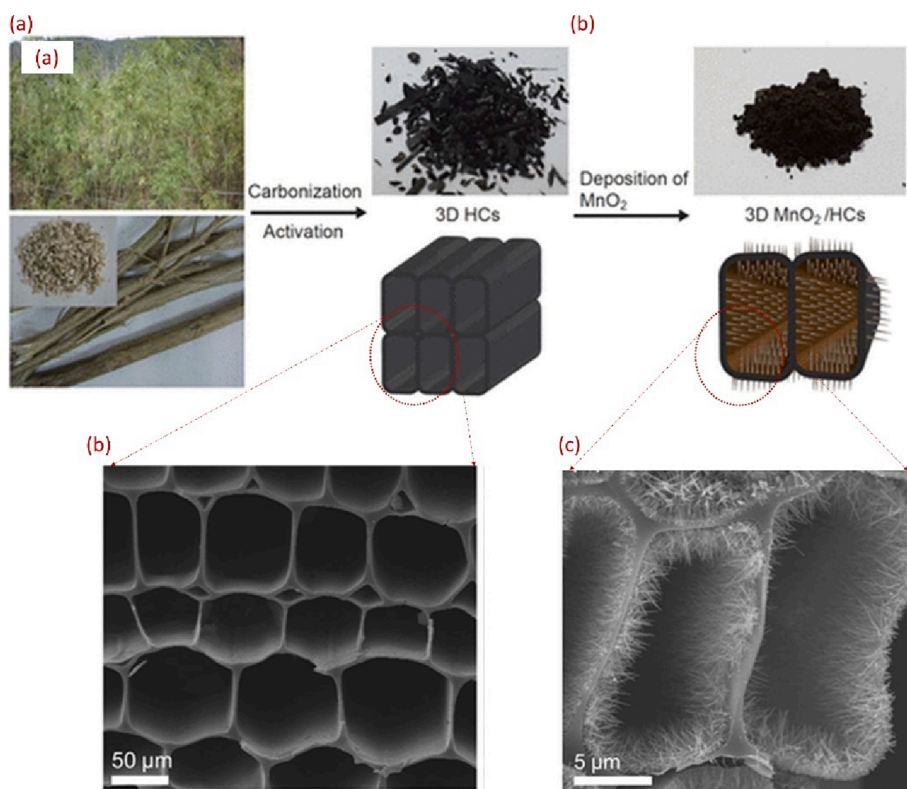


**Fig. 9.** (a) Preparation of hemp-derived carbon nanosheets (CNS), with the lignin (S1) crystalline cellulose (S2) and semi-cellulose (S3) based structural layers, Reprinted with permission from [46] Copyright (2013) American Chemical Society. (b) Ragone chart (energy and power density variation) of CNS obtained at 800 °C evaluated at different temperatures (0–100 °C) Reprinted with permission from [46] Copyright (2013) American Chemical Society.

studies on hemp fibres show that controlled carbonization and steam-activation processing (as shown in Fig. 10a) leads to the development of a honeycomb like 3D macroporous structure (given in Fig. 10b) [47]. Such microstructure could be utilized as a substrate for depositing electroactive pseudocapacitor materials like  $\text{MnO}_2$ ,  $\text{RuO}_2$  etc. for further enhancing the capacitive property of EES devices.  $\text{MnO}_2$  wires growth by the hydrothermal method (as shown in Fig. 10c) on a 3D carbon structure leads to the enhancement ( $\sim$ doubling) of specific capacitance from 163 to 312  $\text{F} \cdot \text{g}^{-1}$  at 25  $\text{mV} \cdot \text{s}^{-1}$  in 1 M  $\text{Na}_2\text{SO}_4$  electrolyte. During charging/discharging measurement, the  $\text{MnO}_2$  wire grown on the honeycomb like 3D structure gave a specific capacitance (340  $\text{F} \cdot \text{g}^{-1}$  and 44  $\text{F} \cdot \text{cm}^{-3}$ ) which is higher than that for  $\text{MnO}_2$  (116  $\text{F} \cdot \text{g}^{-1}$ ) and for 3D honey comb structures (168  $\text{F} \cdot \text{g}^{-1}$ ) [47]. Moreover, the full SC based on  $\text{MnO}_2$ /3D carbon gives an energy density of 33.3  $\text{W} \cdot \text{h} \cdot \text{kg}^{-1}$  and a power

density of 7.8  $\text{kW} \cdot \text{kg}^{-1}$  with 98% retention in capacitance after 3000 cycles [47].

Chemical activation via  $\text{H}_3\text{PO}_4$ ,  $\text{ZnCl}_2$ ,  $\text{KOH}$  may lead to chemical pollution and the presence of impurities. In flax fibre SC fabrication, it was noted that  $\text{CO}_2$  physical activation could provide a lower environmental impact/high purity route to realize activated carbon [50].  $\text{CO}_2$  activation can also lead to the removal of organic impurities and produce a high active surface area with porous carbon electrodes. In a recent work, carbonization at a low temperature of 300 °C for 2 h and  $\text{CO}_2$  activation at 900 °C for 30 h lead to the development of activated carbon from hemp fibres with specific capacitance of 600  $\text{F} \cdot \text{g}^{-1}$  (for an electrode) and 457  $\text{F} \cdot \text{g}^{-1}$  (for full cell), respectively in PVA-KOH electrolyte. The full cell showed an energy density of 25.3  $\text{W} \cdot \text{h} \cdot \text{kg}^{-1}$  at 1  $\text{A} \cdot \text{g}^{-1}$  and a power density of 4320  $\text{W} \cdot \text{kg}^{-1}$  at 10  $\text{A} \cdot \text{g}^{-1}$  with an 85%



**Fig. 10.** (a) Schematic for preparation of 3D  $\text{MnO}_2$  deposited hemp activate carbon (3D  $\text{MnO}_2$ /HC) [47] and (b) shows corresponding 3D HC SEM image with (c) SEM image of 3D  $\text{MnO}_2$ /HC [47]. (a-c) Reprinted (adapted) with permission from [47] Copyright (2017) American Chemical Society.



retention in capacitance after 10,000 charge–discharge cycles [50].

### 3.4. Kenaf fibre based EES

The kenaf fibre is considered as a versatile economic crop (tolerant to environmental stress) for industrial applications including textile fabric, and thermoplastic composites [107]. The high lignin content present in the kenaf, leads to a low ash production and a high quality activated carbon which has potential advantages in conductive frameworks (current collector) for energy storage devices [107]. By activation with different chemicals including  $K_2C_2O_4$ ,  $H_3PO_4$ , etc., [107,108] high quality activated carbon materials have been prepared. For energy storage device fabrication, large surface area, pore size and pore distribution are important and it was found that hierarchical porous carbon materials possess those properties [109]. Such hierarchical porous carbon material structures were prepared from kenaf stems by impregnating different concentrations of Ni ions into kenaf and by hydrochloric acid etching. The SC fabricated using such carbon exhibited energy and power densities of 16.0–12.3 W. h.  $kg^{-1}$  and 200–10 068 W.  $kg^{-1}$  respectively [109].

As discussed previously, 3D structured porous carbon has significant advantages in metal oxide growth/loading. In a recent paper, by using kenaf stems, a 3D porous carbon (3D-PC) was developed as shown in Fig. 11a and was used as a support for  $MnO_2$  loading (as shown in Fig. 11b) by electrodeposition. The developed electrode showed excellent performance for SC fabrication due to the pseudocapacitance contribution from flower-like  $MnO_2$  nanospheres [110]. CV analysis, (as given Fig. 11c) reveals that compared to Pt and 3D-PC based electrodes,  $MnO_2$ /3D-PC electrodes showed outstanding performance at scan rates of 5  $mV. s^{-1}$  [110]. The reported performance of SCs fabricated using  $MnO_2$ /3D-PC is given in the Table 3. In addition to the loading of  $MnO_2$ , the 3D-PC from kenaf fibre was also mass loaded with polyaniline (PANI) [111] and in another work with rGO and PANI [112]. The fabrication steps of kenaf carbon fibre with rGO/PANI are given in Fig. 11d [112]. Furthermore, carbon derived from kenaf fibre was also used for the development of Li-ion capacitors. For this, the kenaf-derived nanoporous activated carbons (KACs) with nanometric hexagonal

micropore structures were prepared by chemical activation with KOH [91]. Such Li ion capacitors exhibited specific capacities of 195  $mA. h. g^{-1}$  with 85% capacity retention rate after 10,000 cycles at 1  $A. g^{-1}$  as presented in Fig. 11e [91].

In section 3 the main methods for electrode fabrication from bast fibres, and its development for energy storage devices are discussed. In EES device fabrication, either porous carbon or conductive polymer coated jute are generally used. As the most abundant natural Fibre, carbon derived from bast leads to a sustainable route to material manufacture for conductive electrodes. The increasing conductivity of carbon through nitrogen doping strengthens its potential utility as an electrode for EES devices. Conductive nanoparticle coating or composite nanostructure coating may also enhance the conductivity and performance of fibre based EES devices. Further studies are required for these Fibres for Li ion storage electrodes, in addition to SCs.

## 4. Future perspective and conclusions

### 4.1. Sustainable materials

The majority of bast fibre based EES devices are based on the activated carbon and its modification with metal oxide, CNTs and conducting polymers. Porous carbon electrodes have been explored for Li ion storage electrodes in a small number of reports and its performances reveals that further modification of the porous electrode with Li-based composite materials will lead to the future development of a full cell. 3D structured carbon generated through chemical or physical activation may be used as a current collector, active electrode in a SC, or anode in a battery. Moreover, currently jute fibre has been modified with conducting polymer for SCs through surface coating. The coating of pseudocapacitive materials, including  $MnO_2$ ,  $RuO_2$  and  $WO_3$  will support the enhancement of the energy storing performance of such SCs. To this end, the bast fibre may be converted to a conductive electrode through conducting polymer coating. The energy storing performance of this electrode may be enhanced through coating with a metal oxide film that could be deposited via electrodeposition [113]. Also surface engineering or modification of materials including graphene or carbon could

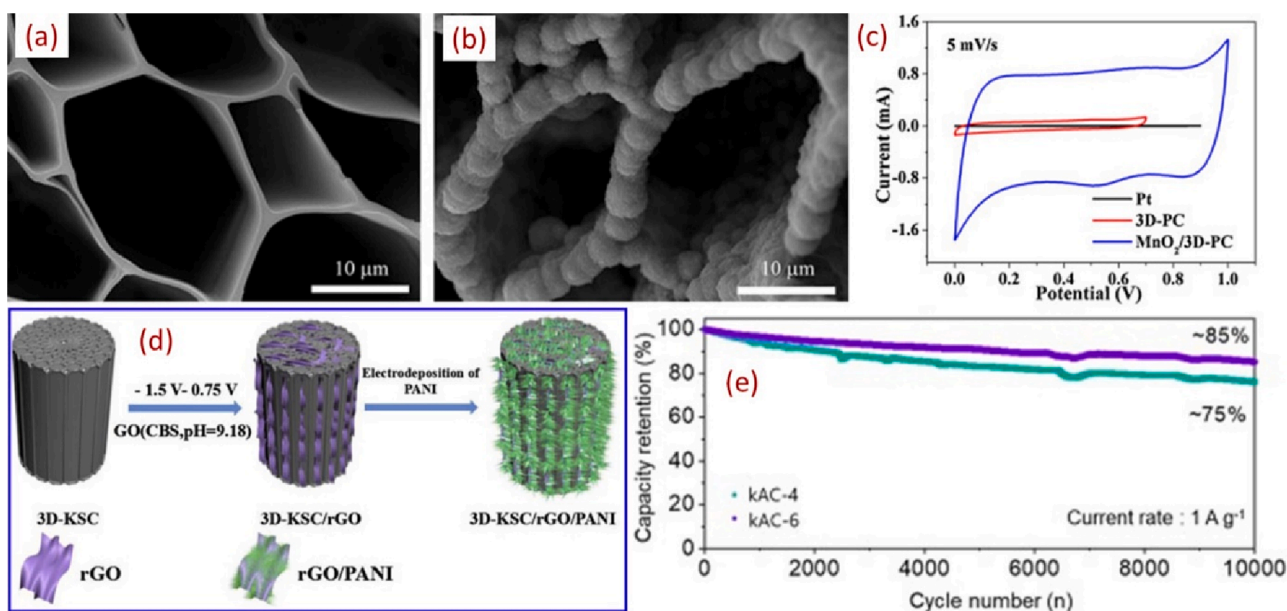


Fig. 11. (a) and (b) SEM image of 3D porous carbon (3D-PC) from kenaf fibre and electrodeposited  $MnO_2$  on 3D-PC. Reprinted from [110] Copyright (2014), with permission from Elsevier, (c) Comparison of CV analysis for Pt electrode with 3D-PC and  $MnO_2$ /3D-PC Reprinted from [110] with permission from Elsevier. (d) schematic of fabrication step of 3D porous kenaf derived carbon (KSC) with coating of reduced graphene oxide (rGO) and PANI Reprinted from [112] Copyright (2018), with permission from Elsevier, (e) long cyclic performances of KAC electrodes with different ratio of KOH for activation (with KOH 1:4 (KAC-4) shows ~75% capacitive retention whereas KOH 1:6 (KAC-6) exhibit ~85% capacitive retention) Reprinted from [91] Copyright (2021), with permission from Elsevier.

enhance the energy storing performances [114].

In addition to active electrodes, the separator is also of significant importance for the development of SCs and batteries. The high tensile strength and high cellulose content of bast fibres show its applicability as a separator after modification of the bast fibre composite. Moreover, the implementation of new biocompatible electrolytes, including gel or aqueous media, needs to be further investigated for improving the operating window of the device and to enhance the life cycle. This will also depend on packaging of the device where soft sustainable material based (e.g., insulative polymers) protective films are further required on alternative to rigid packaging. A summary of bast fibre EES requirement is given in Fig. 12.

#### 4.2. Textile based energy storage devices for wearables

Textile-based electrochemical energy storage devices (TEESDs) including textile-based SCs, batteries, and hybrid supercapacitive battery are ideal for portable and wearable applications [8,113]. As compared to other plastic/polymer substrate-based energy storage devices, the major advantages of the TEESDs are their high flexibility, stretchability, non-toxicity and washability [113,115]. Such TEESDs are adding up to the comforts of the end users and other consumer electronics applications. For the development of sustainable TEESDs, it is essential to consider various components in the fabrication. This includes the substrate, intermediate or protective layer, current collectors, active electrode, electrolyte, conducting cables, soldering materials and packaging material. The major four things about the design of TEESDs include (i) materials, (ii) material processing and fabrication methods, (iii) recycling of the materials, and (iv) performances of the devices. Such TEESD fabrication of the bast fibre-based electrode could be highly useful due to its high performances (as shown in Table 3) and biocompatibility.

According to Table 3, EES based on natural bast fibres will be potential sources for power distribution/management and could be advantageous for reaching future sustainable energy solutions. EESs in pouch and wire/yarn type configurations are the most widely reported in the literature, and are highlighted as the ideal candidates for portable

and wearable applications. Yarn-type configurations made using bast fibres with conducting materials coating may bring additional advantages since their flexibility may allow the wearables to be better adaptable for the wearer. Such device can be directly built into the base fabric (for instance, in the first stages of fabric fabrication, by well-established and high-throughput textile processes). This ensures wearability, breathability, seamless integration, and high functionality, even if stretching or twisting is induced by the wearer, which is not completely guaranteed by pouch-type integration since it is simply an add-on to a surface [116]. Up to now, this has not been implemented in practical applications to power current wearable devices except for LEDs or watches for a short period of time as a proof of concept [24]. For wearable based EES device fabrication, selecting the most appropriate materials, whether substrates, current collectors, active materials, electrolytes, or even the encapsulating layers, should take into consideration their compatibility, electrochemical performance, and EES charge storage mechanism and architectures. Recently, several functional electrolytes have been proposed to monitor the internal conditions and parameters of EES devices [117], and this has inspired worldwide research interest in this field.

#### 4.3. Power management interface and applications

The rapid development of smart low-power electronics, including wearable devices, has unlocked the potential of the usage of EES devices with different energy output ranges (nW- W) and power sources with mechanical flexibility and biocompatibility for implementation on conformable surfaces [118]. Whilst immature as rigid Li-ion batteries, flexible EES devices are starting to make their mark. A recent progress report on 1D SCs [119], for instance, estimates that a Huawei band 2 (1 mW) can be powered by five specific 1D SCs, connected in series, for up to five days. Commercially, this device is sold with a lithium-ion battery that lasts up to 21 days. Enhancing the electrochemical performance of flexible EES devices, including batteries, and SCs is therefore a key future research target to achieve the desired power and energy density, allowing power delivery to both low-power and high-energy-density wearable devices [118,119]. There is so far little work has been carried

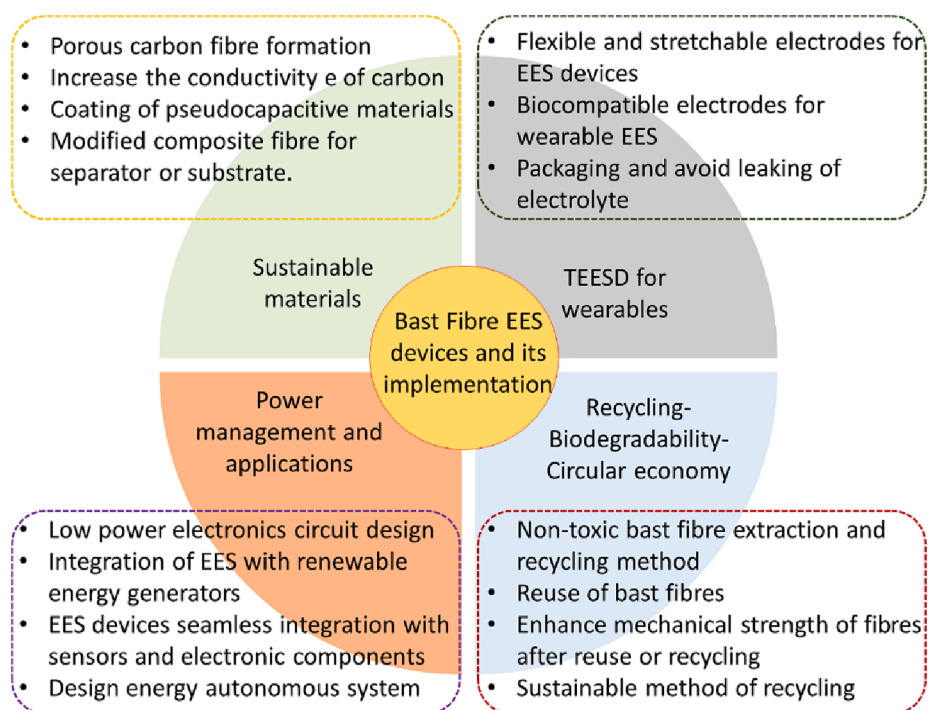


Fig. 12. Future perspective of bast fibre based EES devices and its implementation.

out for power management of bast fibre based EES devices. It is anticipated that improvement in the output power density will be achieved in the next few years. Sufficient improvement would make EES devices not only an output power source but also an input source to harvest energy from renewable resources, i.e. solar, wind, tidal wave, and microwave wave [120–123]. This has opened a vast range of applications that aims to improve the quality of life by using innovative sensor technologies in lifestyle, fitness, healthcare, manufacture, and daily life. Step-by-step flexible EES devices will shift from the prototyping stage to mass fabrication for commercialization, owing to the future development of multidisciplinary fields. Future smart electronics will require their power sources to be more reliable under extreme conditions. To interconnect EES devices to the load, it is required to use efficient power electronic/management interfaces for maximum power transfer. Several electronic power converter topologies such as the standard, multilevel and multiport based structures have been studied over the last decade and this is still a highly active research topic [124].

#### 4.4. Recycling, biodegradability and circular economy

In many cases the materials used may be biocompatible or abundant. However, the method of fabrication or synthesis may be toxic and/or may release large amounts of CO<sub>2</sub>. Similarly, recycling processes at the end of product life may use toxic chemicals. Considering the circular economy, the reusability of utilised materials is essential when considering both fabrication and recycling. All these aspects will strongly influence the performances of the device. Without compromising the energy and power density of the device, the shelf life of the device needs to be enhanced. In this aspect, as this review has highlighted, natural bast fibres based EES devices have significant potential advantages due to their excellent device properties including the development of activated carbon and surface engineered electrodes.

The circular economy concept aims to overcome issues associated with recycling and reusability, and appears to be an inevitable social and economic transition. The concept “Reduce-Reuse-Recycle” aims to limit the impact of our current economic structure, spanning social inequality, depletion of natural resources, and environmental pollution [125,126]. In this context natural bast Fibre is a great choice to use as a biopolymer, that can easily be recycled, reused, or decomposed in a sustainable manner. The sudden rise of the market of natural bast Fibres indicates the global awareness of the use of these materials and their wide range of applications in the green and sustainable field. Due to their recyclability and eco-friendly nature, even after disposal in landfill, natural bast Fibres are exhibiting high levels of interest in both research and industrial fields. Natural bast Fibres may be reprocessed or recycled mainly by “chemical reprocess” or “mechanical reprocess” by the combinations of granulation, grinding, shredding, mixing, melt compounding, extrusion, palletization, compression molding, and injection molding. However, from an industrial point of view the recycling processes should be technically simple and cost-effective.

In chemical recycling methods, the main components from the composite forms of the Fibres are chemically separated and processed either into a different product. On the other hand, by the mechanical recycling processes, used natural bast fibres are mostly retained in their original form, but into a similar product. However, it is important to maintain the basic physical and mechanical properties of the Fibre composites and cost-effectiveness after recycling cycles. The recycling process of the composites also varies depending on the final area of application [127]. Mechanical strength, such as tensile stress–strain, and elasticity of the natural bast Fibres depends on the chemical structure of the cell wall, which is mostly affected during the recycling process. These properties have been shown to both improve and worsen through multiple reprocessing cycles. It has been shown that natural bast Fibre composites may be recycled 5 ~ 8 times until their mechanical performance decreases by up to 15–30% with appropriate coupling agent incorporation during the recycling process [128]. There are also a few

reports where the tensile strength and modulus of the Fibre composites first increased and then decreased with subsequent recycling steps. Some of the reports also show no change in tensile stress after 3–4 recycling cycles [128]. However, recycling processes become complicated when the product from natural bast Fibres are composites with dissimilar materials. It is therefore difficult/expensive to recycle the Fibres from such composites, limiting viability. However, due to international and national legislation, the adoption of circular economy principles is increasing, leading to the predicted widespread future use of natural bast Fibres as prime resources for product manufacturing.

## 5. Conclusions

We have reviewed the fabrication procedures and performance of various natural bast fibres-based energy storage devices. As an environmentally abundant alternative, activated carbon from bast fibres, or the development of new electrodes from bast fibres shows great promise for the fabrication of supercapacitors and/or batteries. Different activation methods, both physical and chemical, used for the preparation of active carbon electrode from biomass of bast fibres have been discussed. The rich fibrous nature of bast materials gives advantages for the electrode development and carbon fibre formation. We noted that the activation methods depend on the formation of pores, pore distribution and shape of the carbon fibres. We highlighted key studies of the improvement of the conductivity and mass loading capacity of natural fibre derived carbon. A detailed comparison on the performance obtained for jute, flax, hemp, and kenaf fibres as EES devices has been reported. It was noted that, a further modification of the material synthesis and enhancement in energy density will lead to the implementation of bast fibre-based energy storage devices into sustainable and clean energy systems, with ease of implementation in the future circular economy being a key differentiator for these materials. In addition to the modification of EES design, we found that an area for significant future study is in power management and system implementation.

## Declaration of Competing Interest

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

## Data availability

Data will be made available on request.

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