High-performance nanostructured bio-based carbon electrodes for energy storage applications

Adel Al Rai · Meltem Yanilmaz

Abstract Polyacrylonitrile (PAN)-based carbon precursor is a well-established and researched material for electrodes in energy storage applications due to its good physical properties and excellent electrochemical performance. However, in the fight of preserving the environment and pioneering renewable energy sources, environmentally sustainable carbon precursors with superior electrochemical performance are needed. Therefore, bio-based materials are excellent candidates to replace PAN as a carbon precursor. Depending on the design requirement (e.g. carbon morphology, doping level, specific surface area, pore size and volume, and electrochemical performance), the appropriate selection of carbon precursors can be made from a variety of biomass and biowaste materials. This review provides a summary and discussion on the preparation and characterization of the emerging and recent bio-based carbon precursors that can be used as electrodes in energy storage applications. The review is outlined based on the morphology of nanostructures and the precursor’s type. Furthermore, the review discusses and summarizes the excellent electrochemical performance of these recent carbon precursors in storage energy applications. Finally, a summary and outlook are also given. All this together portrays the promising role of bio-based carbon electrodes in energy storage applications.
Introduction

Although the year 2020 has witnessed a significant drop in CO₂ emission due to global lockdown measures against COVID-19 pandemic (Le Quéré et al. 2020; Liu et al. 2020e), tremendous efforts are needed to systematically tackle climate change. Depending more on renewable energy sources and shifting toward and relying more on environment friendly solutions, e.g. electric vehicles, is one of the measures toward a sustainable environment. With that, continuing to develop more efficient storage units is becoming the momentum to advance such sustainable technologies (Moriarty and Honnery 2016). Although substantial research and efforts have been made to enhance and develop energy storage units, energy storage is still a barrier against fully benefiting from renewable sources, and a significant increase in capacity is necessary to meet future demands (Leonard et al. 2020).

A conventional energy storage unit is composed of three functional parts: electrodes, liquid electrolyte, and separator. The working principle can be summarized by ions traveling from one electrode to the other passing through the separator during charging and discharging generating energy and power. In the recent literature, rechargeable batteries, such as
lithium-ion batteries (LIBs), and supercapacitors (SCs) are the most developed and researched energy storage devices (Mehtab et al. 2019). LIBs offer high energy density, while SCs offer high power density and having excellent cyclability and stability. In general, the advancement of such energy storage devices can be briefly summarized based on their functional parts as the following: The advancement of liquid electrolytes is mainly related to ionic conductivity and thermal and electrochemical stabilities (Liu and Yu 2019). While the advancement of separators is correlated with ionic resistivity and the overall safety of the unit (Muzaffar et al. 2019; Rai et al. 2021). The electrochemical performance and stability are related to the advance of the electrodes, which are the sole focus of this review.

In energy storage devices, carbon-based electrodes are extensively under research due to their excellent performance. The highly regarded performance is the result of miscellaneous and porous morphologies, ease of modification, and high electrochemical stability and specific capacitance (Endo et al. 2000; Ishikawa et al. 2006; Zhang and Zhao 2009). Carbon-based electrodes have been derived from a verity of petroleum chemicals such as poly(vinyl alcohol) (PVA) (Park et al. 2011), polyethylene oxide (PEO) (Lewandowski et al. 2001), pitch (Meng et al. 2017), polyvinylidene fluoride (PVDF) (Son et al. 2020), and polyacrylonitrile (PAN) (Stojanovska and Kilic 2019). Among them, PAN is the most commonly used precursor. PAN has shown to be an excellent carbon precursor due to its high carbon yield, high quality carbon structure, and formation of wide nanostructures and morphologies. Researchers have managed to synthesize and fabricate novel morphology of nanostructures such as flower particles (Chen et al. 2018b), hollow nanoparticles (Cao and Kruk 2015), ultrafine nanofibers (Lazzari et al. 2007), 2D-nanostructures (Zhong et al. 2014), and other novel nanostructures (Kopeć et al. 2019). Each of which possesses unique surface and physical properties making them ideal as electrodes in energy storage devices. For example, high energy density of 4.03 Wh kg$^{-1}$ with excellent cycling stability over 1000 cycles were recorded for PAN-based cloth electrode in SC (Zheng et al. 2019). While using random PAN-based carbon nanostructures for an electrode in lithium-sulfur battery, 840 mAhg$^{-1}$ was recorded for the initial reversible capacity along with high cycling stability over 150 cycles (Zhang et al. 2019c).

Unfortunately, PAN and most of the other carbon precursors are petroleum chemicals, nonrenewable sources, and of toxic nature to the environment. Therefore, it is imperative to start depending on a new alternative (Fava et al. 2015). For any alternative to take over PAN’s position as an excellent carbon precursor needs not only to sustain all its good advantages, but also needs to offer what PAN could not provide, hence, environmental sustainability, low-cost, and better electrochemical performance. This alternative is rather a collective of renewable and sustainable materials of bio-based origin. Bio-based carbon precursors are environmentally sustainable materials and of low-cost. In fact, the usage of biomass and biowaste materials as carbon electrodes in energy applications falls under the broad dentition of biorefinery (Fava et al. 2015), which one of its aims is converting bio-based resources into value-added products (Cherubini 2010).

To shed a light on the low-cost virtue, a survey on the available market prices of some of bio-based carbon precursors is composed in Table 1. It is clear that bio-bases sources offer a substantial economical advantage in the synthesis and production of carbon electrodes in energy applications. This advantage comes from the fact that most of biomass materials are the byproducts of agriculture (e.g. corn, rice, and wheat straws) or industries (e.g. lignin). Furthermore, most of biowaste food (e.g. rotten fish and shell of shrimps) and plants (e.g. flowers and phoenix tree leaves) are practically free of charge. It is worth noting that, in principle, the carbonization process of bio-based materials is more or less the same with that of PAN. As a result, the processing cost of PAN and bio-based precursors are comparable. Notable efforts to reduce the processing cost and time are evident. For example, increasing the production output of nanofibers using electrically-assisted solution blow spinning (Rai et al. 2020), and reducing carbonization process time by utilizing microwave assisted-hydrothermal (Naghdi et al. 2017) and microwave plasma pyrolysis (Islam et al. 2017) have been reported. As a consequence, bio-based materials are not only low-cost carbon precursors, but also can be processed at least at the same cost of PAN.

Electrochemical performance of biomass- and biowaste-based carbon electrodes is another critical
factor to define the feasibility of bio-based carbon precursors in energy storage applications. Generally, electrochemical performance of carbon-based electrode is correlated with the morphology, porosity and pore size, specific surface area, and doping level. Bio-based carbon electrodes possess unique nanostructures, high specific surface area, and high level of heteroatoms-doping. For some precursors, unique nanostructures are inherited from the biomass materials before carbonization (Wang et al. 2019b). Protein-based carbon (e.g. silk regenerated) and some other biomass-based, e.g. chitin, precursors retain a large amount of nitrogen, oxygen, and sometimes sulfur species after carbonization leading to self-doping (Chen et al. 2018a; Zhou et al. 2019). As a result of these superior physical and chemical features, high-performance electrodes in rechargeable batteries and SCs have been reported as is discussed and summarized in detail in “Bio-based nanostructured carbon materials for high-performance energy applications” section.

Environmentally sustainable biomass- and bio-wastes-based carbon electrodes are excellent options for high-performance energy storage applications. This review provides a summary and discussion on the preparation and characterization of the emerging and recent bio-based carbon precursors that can be used as electrodes in energy storage applications. The review is outlined based on the morphology of nanostructure and the precursor’s type. For each section, preparation protocols and properties are thoroughly discussed. Furthermore, the review discusses and summarizes the excellent electrochemical performance of these emerging carbon precursors in storage energy applications. All this together portrays the promising role of bio-based carbon electrodes in storage energy applications.

Preparation and properties of nanostructured bio-based carbon materials

Particulate nanostructures (0D-nanostructures)

This category encompasses bio-based carbon powders with different morphologies such as nanoparticles, nanosheets, nanoplates, random geometrical nano-sized configurations, or nonporous structure.

Lignin

Lignin, a complex amorphous biopolymer that acts as a binder to provide structural integrity in plants, is the most abundant natural material after cellulose (S. B. Lalvani 2000; Zakzeski et al. 2010). The synthesis of different lignin nanoparticles is an emerging interest (Gonzalez et al. 2017; Ago et al. 2017; Matsakas et al. 2018) but the scope of lignin-based carbon nanoparticles is still rather limited. The reason is that currently most of the synthesized lignin nanoparticles are in colloidal form, which impedes subsequent treatments such as carbonization and

| Carbon precursor                              | Average cost ($/ton) | References                  |
|-----------------------------------------------|----------------------|-----------------------------|
| Corn straw                                   | 29                   | Edwards (2020)              |
| Rice straw                                   | 19.3                 | Delivand et al. (2011)      |
| Wheat straw                                  | 60.4                 | Littlewood et al. (2013)    |
| Chitin (from seafood)                        | 110                  | Yan and Chen (2015)         |
| Kraft lignin                                 | 380                  | Ludmila et al. (2015)       |
| Organosolv lignin                            | 400                  | Ludmila et al. (2015)       |
| High purity lignin                           | 750                  | Ludmila et al. (2015)       |
| Cellulose (from sludge for nanofiber production) | 113              | Jonoobi et al. (2012)       |
| Cellulose (dissolving cellulose for nanofiber production) | 1648            | Jonoobi et al. (2012)       |
| PVA                                           | 2080*                | Echemi (2020)               |
| PAN (used for carbo fiber production)         | 4000*                | Singh Gill et al. (2017)    |

*These prices may change significantly depending on the material grades, origin, and intended application.
activation. The most common configuration of lignin-based carbon nanoparticles is carbon dots forming colloidal suspension (Chen et al. 2016b; Rai et al. 2017; Si et al. 2018; Myint et al. 2018). The reported physical properties, e.g. specific surface area, of these carbon nanoparticles (Yiamsawas et al. 2017) are inferior to conventional activated polymeric-based carbon nanoparticles (Zhao et al. 2015b; Zhou et al. 2015). One report simultaneously managed to carbonize and activate lignin-based carbon nanoparticles with large surface area, 1100 m$^2$/g, however, the obtained nanoparticles showed signs of agglomeration and irregularity in their structure (Hu and Hsieh 2017). As a result of the difficulty of handling colloidal lignin-based carbon nanoparticles, a promising recent study featured the recovery of lignin nanoparticles in solvents using a two-step process (Matsakas et al. 2020); decreasing the solvent concentration and, subsequently, isolating the particles. The rate at which the solvent concentration evaporates was important to obtain good quality particles. In another study, freeze-drying method followed by subsequent stabilization and carbonization was used to overcome the isolation issue, however, the nanoparticles were of irregular shapes (Gonugunta et al. 2012).

Cellulose

Cellulose makes up a large portion of the plant’s interior structure and is the most abundant natural material. Cellulose has been extracted from green plants, e.g. wood pulp. Due to the nature of cellulosic sources, their nanostructures are mainly fibrous and whiskery (see “Cellulose” section), while other particulate structures such as nanoparticles are difficult to obtain. Hence, in limited studies, randomly graphitic nanostructures are the only formation that can be found for limited applications, such as proton exchange membrane fuel cell (Guilminot et al. 2008; Sevilla and Fuertes 2010). This may not just indicate that the cellulose-based carbon nanofibrous is a more common structural configuration than its nanoparticulate counterparts, but maybe also easier to synthesize and apply in a variety of applications.

Chitin

Chitin, an abundant biopolymer with high nitrogen content due to the presence of N-acetyl groups, can be cheaply obtained as a by-product of seafood bio-wastes, for example, shells of crab, shrimp, and lobster (Yan and Chen 2015) and also can be extracted from insects (Zhou et al. 2017). Only a few reports have tackled the synthesis of carbon particulate nanostructures using chitin as a precursor. Elastic nitrogen-doped chitin-based CNFs microspheres were synthesized (Duan et al. 2016). Scanning Electron Microscopy (SEM) and Transmission Electron Microscopy (TEM) were used to capture the marvelous morphology as depicted in Fig. 1. Although the synthesis protocol is complicated and long, the inner structure of the nanofibrous microspheres is outstanding. The CNFs and sphere diameters were measured to be around 24 nm and 39 μm, respectively. The microspheres that were carbonized at 900 °C were partially graphitized and had very high specific surface area and pore volume of 1147 m$^2$/g and 2.12 cm$^3$/g, respectively.

Chitin-based carbon particulate nanostructures could be synthesized in different geometries demonstrating the capability of chitin to be a miscellaneous carbon resource. Another form of chitin carbon particulate nanostructure is carbon quantum dots. In one example, spherical diameters between 2 to 12 nm have been synthesized (Naghdi et al. 2017). Briefly, in a sealed vessel, a predetermined grammage of chitin nanofibers mixed with CaCl$_2$ ethanol solution was subjected to a temperature of 180 °C using one-pot microwave assisted-hydrothermal method. The change of color from colorless to dark was marked as evidence of the conversion to carbon quantum dots. Another example, chitin-based carbon nanosheets have also been synthesized by hydrophobization-induced interfacial-assembly method (You et al. 2017). First, the extracted chitin nanofibers from carb were dissolved in NaOH–urea before drop-wise addition of acrylonitrile that triggered hydrophobization reaction. The reported thickness of the resultant nanosheets was 27 nm and was reduced to around 3.8 nm with lateral size of 10 μm to form graphene-like structure after carbonization at 800 °C. The obtained chitin-based nanosheets carbon had high specific surface area and pore size range of 724 m$^2$/g and 1–40 nm, respectively.

Recent reports have demonstrated the activation with somewhat unconventional materials, e.g. KMnO$_4$ and Hydroxylapatite, that help in increasing specific surface area and nitrogen doping percentage as well
For instance, activation of chitin in hot KMnO$_4$ solution resulted in high specific surface area up to 1941 m$^2$/g at a relatively low carbonization temperature of 800 °C (Wang et al. 2020b). The large surface area was attributed to dismantling hydrogen bonds due to KMnO$_4$ reacting with chitin molecules before carbonization and the decomposition of by-products, e.g. K$_2$CO$_3$, during carbonization. Interestingly, KMnO$_4$ also acted as a template to form nanoporous structures during carbonization. Nonetheless, one of the drawbacks of chitin is its limited carbon yield, for instance, at a carbonization temperature of 800 °C carbon yield is only 10.5% (Wang et al. 2020b).

Protein.

Proteins are widely abundant bio-based materials and rich of nitrogen species (Li et al. 2013; Demir et al. 2018). Due to the nature of extracting protein precursors, the conventional spherical nanoparticle structure is a rare configuration, while nanoporous powder and nanoplates are more common configurations (Demir et al. 2018). Carbonized protein-based nanoporous
carbon precursors possess excellent physical properties (Li et al. 2013). Some of the promising protein-based carbon precursors are egg, silk, fish, tofu, and some livestock feed such as Lemna minor, and enoki mushroom.

High sources of protein such as activated egg white and eggshell were carbonized and resulted in nanoporous carbon powders (Li et al. 2016a; Zhang et al. 2019b). The nanoporous powders of egg white and eggshell have high specific surface area of 2268 and 1572 m$^2$/g, respectively, and pore sizes smaller than 10 nm. Silk is another good candidate that serves as a protein source. Silk protein-based carbon nanoplates were synthesized from cocoons by, briefly, extracting silk fibroin from cocoons and dissolving it in lithium bromide and the mixture was casted to form a film (Yun et al. 2013). The as-casted film was treated with KOH and carbonized at a temperature of 800 °C to form nanoplates of lateral size around 5 μm and thickness of only a few nanometers. These nanoplates are physically similar to graphene and have similar Raman peaks ratio as well. The specific surface area of the activated nanoplates is substantial, 2557 m$^2$/g. Other protein-rich biowastes, e.g. fishes (Guo et al. 2017), have also been converted to nanoporous carbon materials. For example, a fish activated with ZnCl$_2$ and carbonized at 900 °C resulted in nanoporous carbon content with high specific surface area of 850 m$^2$/g and pore size of around 2.5 nm. Other promising protein-based carbon sources are tofu, livestock feed, e.g. Lemna minor, and enoki mushroom (Guo et al. 2015, 2016; Lee et al. 2017, 2018).

Others

Unconventional pristine and composite renewable bio-based, e.g. food and other bio-based wastes, have successfully been used as carbon precursors. Such materials possess high specific surface area and unique morphology that make them attractive carbon precursors for researchers to unravel their potential and find methods to process them. Food wastes, e.g. walnut and coconut shells, potatoes, and tea waste, are important classes of biowaste to obtain high quality carbon precursors (Sun et al. 2013; Long et al. 2015; Wu et al. 2015; Qu et al. 2015; Cao et al. 2019; Shang et al. 2020; Khan et al. 2020). Carbonized alkali-treated wheat flour and reduced MnO$_2$ nanowires-like composite has been simply synthesized in one-pot but with complex inner honeycomb-like nanoporous structure as illustrated in Fig. 2 (Wu et al. 2015). The unique interconnected honeycomb-like morphology was thought to be as a result of the synergetic role KOH played as a template and as an activation agent. Nitrogen-doped sulfur and gelatin-based carbon composite with ultrahigh surface area and total pore volume, 2893 m$^2$/g and 2.8 cm$^3$/g, respectively, were also synthesized by a series of chemical treatments (Qu et al. 2015). The nitrogen-doping was the result of the rich nitrogen species in gelatin while the high surface area and large pore volume were the results of physical activation using KOH. Also, impressively, activated carbonized porous walnut shells and tea waste with high specific surface areas of 3577 and 1610 m$^2$/g, respectively, were synthesized (Khan et al. 2020; Shang et al. 2020).

A variety of biomass sources, e.g. flowers, bagasse, have also been utilized as nanoparticulate carbon precursors (Chen et al. 2016a; Li et al. 2016b; Gao et al. 2017a; Liu et al. 2017a; Zhu et al. 2020b; Wan and Hu 2020). For example, seed-free willow catkin flower has been converted to nitrogen-doped carbon by pyrolysis after being treated with KOH (Li et al. 2016b). The reported morphology consisted of graphene-like nanosheets with high specific surface area of 1533 m$^2$/g doped with heteroatoms nitrogen and sulfur. The carbonization of Perilla frutescens leaves, a common food plant in Southeast Asia, at 700 °C resulted in graphene-like nanosheets with high heteroatoms self-doping of oxygen and nitrogen at around 18.8 and 1.7%, respectively (Liu et al. 2017a). However, exhibited a somewhat moderate specific surface area of 655 m$^2$/g.

Nature biowastes, e.g. plant leaves, bamboo fiber, wheat straw, and dead ants, are yet another important class as nanoparticulate carbon precursors due to their abundance in nature (Du et al. 2019; Ji et al. 2020; He et al. 2020). The resultant activated graphene-like nanosheets of phoenix leaves, bamboo fiber, and wheat straw resulted in high specific surface area of 2208, 2561, and 2560 m$^2$/g, respectively (Du et al. 2019; Ji et al. 2020; He et al. 2020). Another interesting graphene-like nanosheets carbon doped with three heteroatoms, nitrogen, oxygen, and sulfur, was derived from dead ants (Zhao et al. 2018a). This ant-based carbon powder possessed ultrahigh specific surface area of 2650 m$^2$/g with mesopore size range between 2 to 6 nm.
As summarized in Table 2, unconventional biomass and biowaste materials are showing to have high value as carbon precursors due to their distinct morphology and superior physical properties. The abundance of these renewable precursors for nanoparticulate carbon materials allows them to replace their conventional synthetic counterparts.

1D-nanostructure

This category is composed of bio-based materials that have large length-to-width ratio such as nanofibers, nanorods, nanoneedles, etc.

Lignin

Lignin-based carbon nanofibers (LCNFs) offer an important renewable alternative to PAN-based CNFs due to lignin’s abundance and its relatively high carbon yield. There are two main approaches to obtain CNFs using lignin as a precursor, and both approaches involve electrospinning method. The first approach uses lignin as the only precursor to produce nanofibers, while the other approach, more common, uses lignin with other auxiliary polymers to form a spinnable blend. In the first approach, a high concentration of lignin is required (Lallave et al. 2007; Ruiz-Rosas et al. 2010). Usually, a high concentration solution is not considered as a proper solution in electrospinning due to the rapid solvent evaporation that eventually leads to clogging. Therefore, as a remedy to this issue, using a co-axial spinneret, allows for an extra amount of solvent to be supplemented to the polymer jet to compensate for the rapidly evaporating solvent. Co-axial electrospinning, thus, offers a solution to produce CNFs processed from pristine lignin sources.
| Carbon precursor | Additive/activation agent | Doping conditions | $I_D/I_G$ | $d_{(002)}$ (nm) | Specific surface area (m$^2$/g) | Total pore volume (cm$^3$/g) | Pore size (nm) | References |
|------------------|---------------------------|------------------|-----------|-----------------|-------------------------------|-----------------------------|---------------|------------|
| Kraft lignin     | –/–                       | None             | 8         | –               | 552                           | 0.3                         | 14.8          | Yiamsawas et al. (2017) |
| Alkali lignin    | –/KOH                     | None             | –         | –               | 483                           | 0.3                         | 0.6–1.4       | Hu and Hsieh (2017) |
| Alkali lignin    | –/NaOH                    | None             | –         | –               | 1847                          | 1.1                         | 0.6–1.4       | Hu and Hsieh (2017) |
| Chitin (Cicada slough) | –/KOH               | Self-doping      | 1.5       | 33.8            | 2097                          | 1.09                        | 1.5           | Zhou et al. (2017) |
| Chitin           | –/KMnO$_4$                | Self-doping      | 1.4       | 10.6            | 1488                          | 1.29                        | 3.5           | Wang et al. (2020b) |
| Chitin (N-acetylglucosamin) | –/Hydroxylapatite | Self-doping      | High      | –               | 756                           | 2.34                        | 0.7–2.1       | Chu et al. (2020) |
| Protein (Eggshell) | –/KOH                   | Self-doping      | 7.5       | 7.6             | 1572                          | –                           | 1–3           | Zhang et al. (2019b) |
| Protein (Tofu)   | –/KOH                     | None             | –         | –               | 2960                          | 1.47                        | 2.0           | Lee et al. (2017) |
| Walnut shells    | –/KOH                     | None             | –         | –               | 3577                          | 2.19                        | 3–7           | Shang et al. (2020) |
| Tea waste        | –/KOH                     | None             | –         | –               | 1610                          | –                           | 6.0           | Khan et al. (2020) |
| Bagasse          | –/KOH                     | Self- and external-doping | –      | 15.9            | 326                           | 0.42                        | –             | Wan and Hu (2020) |
| Leaves (Phoenix) | K$_2$FeO$_4$/K$_2$FeO$_4$ | Self- and external-doping | 0.8 | 6.7             | 2208                          | 1.27                        | 2.3           | He et al. (2020) |
| Bamboo fiber     | K$_2$Fe(CN)$_6$/KOH & thiourea | Self- and artificial-doping | 3.2 | 6.1             | 2561                          | 1.34                        | < 10          | Ji et al. (2020) |

* Table 2 Comparison of physical properties of different biomass- and biowaste-based carbon particulate nanostructures. *
However, coaxial electrospinning lacks simplicity and requires extreme care to balance the flow rate of both spinnerets during electrospinning. It is worth noting that conventional electrospinning, one spinneret model, has been reported for the production of nanofibers of pristine lignin solution followed by conversion to LCNFs (Lallave et al. 2007; Ruiz-Rosas et al. 2010; García-Mateos et al. 2018). However, other researchers have failed to reproduce similar results (Dallmeyer et al. 2010, 2014) and have concluded that the system is impractical for the production of LCNFs. One group attempted spinning different technical lignins, namely softwood kraft lignin, hardwood kraft lignin, and sulfonated kraft lignin, using one-spinneret electrospinning but failed to produce any fibers regardless of the solution concentration (Dallmeyer et al. 2010). They only managed to produce beaded fibers at a concentration higher than 50% which would not be suitable for long production process due to eventual clogging.

The second approach uses traditional electrospinning by mixing lignin with some auxiliary polymers to form a blend solution that is easily spinnable compared with the poorly spinnable pristine lignin solution. It is important to note, however, that the use of synthetic polymers defeats the purpose of using lignin as a friendly environmental precursor to produce CNFs. This issue has been tackled by minimizing the amount of percentage of the auxiliary polymeric (Ding et al. 2016), e.g. using lignin in higher percentages compared to the auxiliary polymer, or by using water soluble polymers, somewhat friendlier options, e.g. PVP and PVA. As a result, blending has become a much common approach to produce LCNF using conventional electrospinning rather than using the complex coaxial electrospinning setup.

Table 3 summarizes the most common auxiliary blends, solvents, additives and enhancers, and carbonization conditions used to prepare LCNFs using electrospinning method. Lignin has been blended with a variety of polymers in order to enhance the spinnability of lignin to produce nanofibers and subsequently convert them into LCNFs. PAN is the most common auxiliary polymer thanks to its high carbon yield and mechanical properties (Ding et al. 2016). Besides PAN (Ruiz-Rosas et al. 2010; Choi et al. 2013; Xu et al. 2013, 2014; Ding et al. 2016; Dalton et al. 2019; Jayawickramage et al. 2019; Demiroğlu Mustafov et al. 2019; Dai et al. 2019; Zhang et al. 2020b; Du et al. 2020a), many other polymers have also been used as auxiliary polymers to enhance lignin spinnability, such as PVP (Ma et al. 2018; Cao et al. 2020), PVA (Ago et al. 2012; Lai et al. 2014; Ma et al. 2016, 2019; Zhao et al. 2018b; Jayawickramage and Ferraris 2019; Roman et al. 2019), PEO (Dallmeyer et al. 2010; Hu and Hsieh 2013; Cho et al. 2019; Du et al. 2020b), TPU (Culebras et al. 2019), and PLA (Culebras et al. 2019). Worth reporting that, to some less extent instead of blending, direct synthesize of lignin and PAN copolymer has been reported as a precursor of LCNFs for energy applications (Youe et al. 2016, 2018).

To further enhance the lignin/polymer blend spinnability in electrospinning, the literature demonstrates a wide variety of materials as potential blend enhancers. Interestingly, nanocrystalline cellulose as an additive has been used to enhance the spinnability of lignin/PEO blend by controlling the molecular orientation of lignin during electrospinning (Cho et al. 2018, 2019). Non-precious metals have been used as a catalyst to increase the lignin yield but also showed notable enhancement in the spinnability of lignin. For instance, the addition of 10 wt.% of Ni ions to organosolv lignin blend increased the light lignin fragments yield to 87%, and enhanced the solution spinnability during electrospinning (Du et al. 2020b). As summarized in Table 3, other enhancers reported are platinum acetylacetonate, methylene diphenyl diisocyanate, graphene nanosheets, and butyric anhydride (Ruiz-Rosas et al. 2010; Ding et al. 2016; Culebras et al. 2019; Dai et al. 2019).

Depending on the starting materials, it seems that lignin/polymer blend tends to have higher carbon yield than the pristine polymers. Though PAN has the highest carbon yield among synthetic polymers, pristine lignin and lignin/PAN blend results in even higher LCNFs yield (Xu et al. 2014; Ding et al. 2016). The final stabilization and carbonization temperatures are more crucial than heating rate and holding time to determine the final carbon yield. A recent study showed that the final stabilization temperature of lignin-based nanofibers has a much more effect on carbon yield compared with heating rate and holding time combined (Cho et al. 2019). They showed that after stabilization at 200, 230, 250, and 280 °C the yield decreased to around 87, 82, 78, and 65%, respectively. After carbonization at 1000 °C the corresponding carbon yield stood at 38.4, 44.4, 47.9,
Table 3  Process conditions and protocol of lignin derived CNFs using electrospinning

| Carbon precursor | Materials processing conditions | Carbon conversion conditions | Final carbon yield (%) | References          |
|------------------|---------------------------------|-----------------------------|------------------------|---------------------|
|                  | Catalyst/active material/ enhancer | Solvent | Auxiliary polymer | Lignin/ polymer ratio | Blend concentration (wt.%) | Viscosity (Pa.s) | Stabilization | Carbonization |                   |
| Alkali lignin    | KOH                             | Water  | –               | 20                     | –                   | Nitrogen @105 °C @10 °C/min for 0.5 h | Nitrogen @900 °C @10 °C/min for 0.5 h | 18.2        | Hu and Hsieh (2017) |
| Alcell lignin    | –                               | Ethanol | –              | 50                     | 0.35–0.45           | Atmosphere @200 °C @0.25 °C/min for 24 h | Nitrogen @900 °C @10 °C/min for 0.5 h | 31.6        | Lallave et al. (2007) |
| Alcell lignin    | –                               | Ethanol | –              | 50                     | –                   | Atmosphere @200 °C @0.05 °C/min for 36 h | Nitrogen @900 °C @10 °C/min for 0.5 h | 39          | Ruiz-Rosas et al. (2010) |
| Alcell lignin    | Platinum acetylacetonate        | Ethanol | –              | 50                     | –                   | Atmosphere @200 °C @0.05 °C/min for 36 h | Nitrogen @900 °C                  | 38.7        | Ruiz-Rosas et al. (2010) |
| Organosolv lignin | Ni ions                         | DMF    | PEO            | 95/5                   | 25                  | Atmosphere @250 °C @1 °C/min for 1 h | Nitrogen @900 °C @5 °C/min for 1 h | –           | Du et al. (2020b) |
| Alcell lignin    | Nitrogen doping                 | DMF    | PEO            | 90/10                  | ~ 20                | Atmosphere @200 °C @1 °C/min for 2 h | Nitrogen @900 °C @10 °C/min for 2 h | –           | Wang et al. (2013) |
| Low sulfonate alkali lignin | NaOH and KOH                  | Water  | PEO            | 90/10                  | 10                  | Atmosphere @105 °C @10 °C/min for 0.5 h | Nitrogen @850 °C @10 °C/min for 2 h | –           | Hu and Hsieh (2013) |
| Softwood kraft lignin | NCC                            | DMF    | PEO            | 27wt.%/ 1wt.%          | 28                  | Atmosphere @230 °C @5 °C/min for 0.5 h | Nitrogen @1000 °C @10 °C/min for 1 h | 44.4        | Cho et al. (2019) |
| Softwood kraft lignin | NCC                            | DMF    | PEO            | 27wt.%/ 1wt.%          | 28                  | Atmosphere @230 °C @5 °C/min for 0.5 h | Nitrogen @1000 °C @10 °C/min for 1 h | 47.9        | Cho et al. (2019) |
| Softwood kraft lignin | NCC                            | DMF    | PEO            | 27wt.%/ 1wt.%          | 28                  | Atmosphere @230 °C @5 °C/min for 0.5 h | Nitrogen @1000 °C @10 °C/min for 1 h | 49.1        | Cho et al. (2019) |
| Carbon precursor | Materials processing conditions | Carbon conversion conditions | Final carbon yield (%) | References |
|------------------|-------------------------------|----------------------------|------------------------|------------|
| Ethanol-soluble lignin | – DMF PAN 85/15 20 0.144 | Atmosphere @220 °C @0.2 °C/min for 6 h Nitrogen @1000 °C @4 °C/min for 4 h | ~ 51 | Du et al. (2020a) |
| Tetrahydrofuran-soluble lignin | – DMF PAN 85/15 20 0.177 | Atmospheric @220 °C @0.2 °C/min for 6 h Nitrogen @1000 °C @4 °C/min for 4 h | ~ 53 | Du et al. (2020a) |
| N. Enzymatic hydrolysis lignin | – DMF PAN 60/40 12 – | Atmospheric @250 °C @1 °C/min Nitrogen @800 °C @10 °C/min for 1 h | – | Zhang et al. (2020b) |
| Organosolv Lignin | – DMF PAN 70/30 15 – Atmospheric @200 °C @0.1 °C/min for 12 h Nitrogen @900 or 1100 °C @5 °C/min for 1 h | Dalton et al. (2019) |
| Enzymatic hydrolysis | Graphene nanosheets DMF PAN 50/50 20 – Atmospheric @260 °C @0.5 °C/min for 1.5 h Nitrogen @1400 °C @5 °C/min for 1 h | Dai et al. (2019) |
| Organosolv lignin | – DMF PAN 50/50 20 ~ 1.4 Atmospheric @200 °C @0.2 °C/min for 12 h Nitrogen @1000 °C @5 °C/min for 0.5 h | Ding et al. (2016) |
| Organosolv lignin | Butyric anhydride DMF PAN 50/50 20 ~ 2.2 Atmospheric @200 °C @0.2 °C/min for 12 h Nitrogen @1000 °C @5 °C/min for 0.5 h | Ding et al. (2016) |
| Poplar sawdust lignin | PMMA DMF PVP – – – Atmospheric @300 °C @1 °C/min Nitrogen @800 °C for 2 h | Cao et al. (2020) |
| Carbon precursor     | Materials processing conditions | Carbon conversion conditions | Final carbon yield (%) | References                  |
|---------------------|---------------------------------|------------------------------|------------------------|-----------------------------|
|                     | Catalyst/active material/ enhancer | Solvent | Auxiliary polymer | Lignin/polymer ratio | Blend concentration (wt.%) | Viscosity (Pa.s) | Stabilization | Carbonization |                           |
| Kraft lignin        | –                               | Water  | PVA             | 70/30                | 12                          | –               | Atmosphere @100 °C | Argon @1200 °C         | –                          | Lai et al. (2014)    |
| Alkali lignin       | –                               | Water  | PVA             | 80/20                | 24                          | 0.48            | Atmosphere @250 °C | Nitrogen @1000 °C      | 30–35                      | Jayawickramage and Ferraris (2019) |
| Alcell organosolv lignin | MDI                              | DMF    | TPU             | 80/20                | 20                          | –               | Atmosphere @150 °C | Nitrogen @900 °C       | 47.5                      | Culebras et al. (2019) |
| Alcell organosolv lignin | MDI                              | THF/ DMF | PLA             | 80/20                | 20                          | –               | Atmosphere @150 °C | Nitrogen @900 °C       | 32.7                      | Culebras et al. (2019) |
nd 49.1%, respectively. Although this suggests that the higher the stabilization temperature the higher the carbon yield, when stabilization temperature reached 300 °C the carbon yield, at the same carbonization temperature (1000 °C), decreased to 44.8%. Therefore, the optimum stabilization temperature was around 280 °C. Stabilization process not only help increase carbon yield but also avoid fusion of the nanofibers (Cho et al. 2018; Roman et al. 2019), by cross-linking to increase the softening temperature of lignin nanofibers (Kadla et al. 2002; Luo et al. 2011; Chatterjee and Saito 2015). By monitoring the carbonization temperature, the final carbon yield could be controlled for pristine lignin-based and lignin/polymer blended LCNFs. For instance, pristine Alcell lignin nanofibers yielded LCNFs between 47.8 and 36.1% at carbonization temperatures of 600 to 1000 °C, respectively (Ruiz-Rosas et al. 2010), and alkali kraft lignin blended with PAN (1:1) yielded LCNFs between 51.2 and 39.8% at carbonization temperatures of 600 to 950 °C, respectively (Xu et al. 2014). It is worth noting that only a few studies reported carbon yield higher than 50% using ethanol-soluble and tetrahydrofuran-soluble lignins at high carbonization temperatures (Du et al. 2020a). According to Table 3, it could be concluded that carbon yield of lignin-based materials is in the range between 30 to 49%, and, strictly speaking, has a rough average yield of around 42%.

LCNFs, without post- or pre-treatments, already possess somewhat high specific surface area and pore volume (Wang et al. 2013; Ma et al. 2018). These are attributed to the removal of volatile substances during carbonization, such as oxygen which is present at high content on lignin fibers after stabilization (Brodin et al. 2010; Ruiz-Rosas et al. 2010; Baker et al. 2012; Yun et al. 2019). It is worth noting that lignin of lower molecular weight might play a crucial role in the
formation of pores after carbonization (Jeon et al. 2015). Furthermore, specific surface area, pores size, and pore volume of traditional CNFs can be enhanced using chemical agents such as KOH, NaOH, H₃PO₄, K₂CO₃, Na₂CO₃, and ZnCl₂ (Jiang et al. 2002; Gryglewicz et al. 2005; Mitani et al. 2005; Kim et al. 2007; Im et al. 2009). Notably, KOH and NaOH are the common agents to treat LCNFs (Hu and Hsieh 2013; Li et al. 2014; Dai et al. 2019). Physical post-treatment methods, include air plasma, a mixture of CO₂ and N₂ gas, and N₂ gas, were also applied to increase surface area and porosity of LCNFs (Wang et al. 2013; Jayawickramage and Ferraris 2019; Zhang et al. 2020b). On the other hand, pre-treatment methods include the incorporation of porogen in the spinning solution (Ma et al. 2018). Surface treatment and activation of LCNF incorporated with graphene nanosheets were carried out by dispersing the CNFs into KOH solution using ultrasonic treatment (Dai et al. 2019). Graphene nanosheets played an agent role to induce heteroatoms of N and S in LCNF and increase surface area. Furthermore, lignin/polymer blend section does not just affect the spinnability during electrospinning, but also affects the final morphology of LCNF. Depending on the chosen polymer, solution viscosity and electrical conductivity vary that in turn the nanofiber diameter could either increase or decrease. Moreover, if the selected polymer is miscible with lignin, e.g. TPU, the resulted LCNFs are smooth and of low pore volume, e.g. 0.01 cm³g⁻¹, however; if the selected polymer is immiscible with lignin, e.g. PLA, the resulted LCNFs contain pores and have high pore volume, e.g. 0.31 cm³g⁻¹ (Culebras et al. 2019).

Complex carbon-based morphological nanostructures using lignin have been reported. A simple but interesting approach demonstrates the fabrication of a composite LCNF@SnO₂ as hollow multichannel nanofibrous powder using electrospinning method has been reported (Cao et al. 2020). First, at different mass ratios, 1:9, 3:7, 5:5, 7:3 and 9:1, lignin and PMMA were mixed in a DMF solution to which a predetermined amount of PVP and SnCl₂·H₂O were added. The final mixture was used to prepare...
nanofibers using electrospinning. After acid treatment and carbonization of the as-spun nanofibers, LCNF@SnO$_2$ composite as powder was prepared. The reason for obtaining the carbonized composite in powder from perhaps because of the brittleness of the resulted composite. PVA and PMMA were used as spinning auxiliary polymer and as a sacrificial phase material to create multichannel nanofibers, respectively. When the amount of lignin was the smallest (lignin to PMMA 1:9), the nanofibers formed almost a single hollow structure as shown in Fig. 3a, b. As the lignin amount was increased, smaller multiple hollow channels started to form up until solid nanofibers were formed (lignin to PMMA 9:1) shown in Fig. 3c–j. Specific surface area and total pore volume, 659 m$^2$/g and 0.56 cm$^3$/g, respectively, were the highest for multichannel nanofibers (lignin to PMMA 5:5). In addition, with the increase in the lignin amount, the nanofiber diameter decreased from 310 to 210 nm. Lignin offers an alternative green option to PAN to produce LCNFs with tunable morphology.

**Cellulose**

Self-assembly or bottom-up method of cellulose nanofibers is possible by a series of chemical pretreatments, controlling the initial cellulose concentration, and introducing freeze-drying (Han et al. 2013). The process, however, is long and somewhat complicated. Therefore, electrospinning, top-down method, is an attractive and simple method. Due to the poor spinnability of pristine cellulose, electrospun cellulose nanofibers are prepared using cellulose acetate (CA) dissolved in acetone, or a mixture of acetone/DMAc, or a mixture of acetone/DMF/trifluoroethylene. The as-spun membrane can be deacetylated by immersing CA nanofibers in alkaline solutions, i.e. NaOH at room temperature (Deng et al. 2013; Li et al. 2019b; Liu and Hsieh 2002, 2003; Lu and Hsieh 2010; Cai et al. 2015), or NaOH at elevated temperature (Ma et al. 2005), or NH$_4$OH, mixture of NH$_4$OH and NH$_4$Cl (Kuzmenko et al. 2014), or KOH (Son et al. 2004). The salts can be dissolved in ethanol, water, a mixture of ethanol and water, or a mixture of acetone and water, or a mixture of acetone and water. The deacetylated cellulose nanofibrous membranes tend to have a lower average fiber diameter, fiber distribution, and pore size compared with as-spun CA nanofibers (Deng et al. 2013). In addition, the deacetylation process conditions, i.e. salt concentration, mixture ratio, temperature, and immersing duration, affect the final morphology, surface area, surface roughness, and carbon yield of CNFs. For example, cellulose nanofibers immersed in low concentrated NaOH–ethanol or water solutions resulted in rather film-type structure after carbonization (Cai et al. 2015). While, cellulose nanofibers immersed in NaOH–low ethanol/water ratio solution, after carbonization, resulted in interbonded fused nanofibers due to diffusion inability of the mixture causing unhydrolyzed spots. Electrospun polypyrrole-coated cellulose-based CNFs deacetylated in 0.1 M NaOH-ethanol solution and carbonized at 850 °C resulted in nitrogen-doped interconnected network with a specific surface area of 501 m$^2$/g and pore size of 2.7 nm as shown in Fig. 4 (Cai et al. 2015). As Fig. 4 shows, polypyrrole-coated cellulose-based CNFs have rougher surface area than pristine cellulose-based CNFs. The rougher surface was the resultant of the high specific surface area, while nitrogen species were the results of the polypyrrole-coated layer. EDS results in Fig. 4d shows the nitrogen content on the coated nanofibers.

Carbon content in cellulose is around 44%, which is relatively low compared with other carbon precursors, and the final carbon yield after carbonization is only between 10–15%. This low-carbon yield issue was addressed in a two-decade old study by suggesting the impregnation of sulfuric acid to increase the yield (Kim et al. 2001). Sulfuric acid acted as dehydrating catalyst to remove oxygen atoms in form of water instead of CO and CO$_2$ gases that led to smaller loss of carbon atoms during carbonization. At optimum does of sulfuric acid, ~ 6 wt.%, carbon yield reached 38%, which suggested loss of carbon atoms to be around 6% only. To further improve on this, a much recent study, improved the carbon yield simply by altering the composition of the deacetylated solution, NH$_4$OH, with the inclusion of NH$_4$Cl (Kuzmenko et al. 2014). Subsequently, without washing, the deacetylated cellulose nanofibers were carbonized. NH$_4$Cl offered thermal stability and increased carbon yield from 13 to 20%.

Cellulosic nanowhiskers are nanorod-like structure with a diameter between 10 to 30 nm, and unlike cellulosic nanofibers, they do not form interconnected nanofibrous structure. The art of production of cellulosic nanowhiskers (nanorod-like) and the effect of processing conditions can be found in a review that...
provides a thorough comprehension on the topic (Eichhorn 2011). Briefly, depending on the initial cellulose source, nanowhiskers can be prepared using acid hydrolysis method. By controlling processing conditions, e.g. initial source, hydrolysis duration, temperature, and polydispersity, the final morphology and physical properties can be tuned (Eichhorn 2011; Haafiz et al. 2014). Moreover, cellulose-based carbon nanowhiskers, widely referred to as carbon nanoneedles, can be prepared by hydrolysis of microcrystalline cellulose using sulfuric acid at temperature of 45 °C for 3 h followed by thermal stabilization and carbonization at 240 °C and 1200 °C, respectively (Cho et al. 2015). Although some studies show the potential of manipulating the structure of cellulose-based carbon nanoneedle to develop nitrogen-enriched (Silva et al. 2012) and copper, nickel, or iron-doped carbon nanoneedles (Araujo et al. 2016) or form composite (Silva et al. 2015), there is still more work to do to study and improve the current state of cellulosic-based carbon nanoneedles.

The cellulosic part of plants offers a solution to produce cellulose acetate nanofibers using electrospinning, while bacterial cellulose (BC), produced by bacterial fermentation, offers an alternative fast route without the need for electrospinning, due to its readily ultrafine nanofibers. BC nanofibers form 3D interconnected structure with a typical diameter of around 50 nm. Also, BC is distinguished against plant cellulose by its high chemical purity, crystallinity, mechanical property, degree of polymerization, and self-assembly (Shoda and Sugano 2005; Huang et al. 2014). Nonetheless, before carbonization, the network structure of bacterial cellulose pellicles needs to be preserved by subjecting it to freeze-drying (Lee et al. 2013; Yu et al. 2014; Jiang et al. 2016). Although some studies show the potential of manipulating the structure of cellulose-based carbon nanoneedle to develop nitrogen-enriched (Silva et al. 2012) and copper, nickel, or iron-doped carbon nanoneedles (Araujo et al. 2016) or form composite (Silva et al. 2015), there is still more work to do to study and improve the current state of cellulosic-based carbon nanoneedles.

Table 4 summarizes different process conditions and protocol of cellulose derived CNFs. It can be seen that each source of cellulose retains a unique preparation protocol providing suitable and applicable options for industrial scaling.

Chitin

Depending on the source of chitin, chitin nanofibers are extracted by a series of chemical and mechanical treatments with a diameter of around 20 nm. For example, using crab shells, chitin is extracted by treating the shells with NaOH and HCl for few days before being subjected to ethanol for few hours. Grinder treatment is followed to obtain non-aggregate nanofibers (Ifuku and Saimoto 2012). Due to the nanofibrous nature of chitin and its high nitrogen content, nitrogen-doped chitin-based CNFs (CCNFs) can be directly prepared from pure chitin without further subsequent activation. However, CCNFs do not sustain a free-standing membrane but rather nanofibrous powder.

Nitrogen-doped CCNFs have been prepared by thermal stabilization at 300 °C for 1.5 h followed by carbonization at 500, 600, 700, 800, and 900 °C (Hao et al. 2018b). Result demonstrated that as the temperature increased from 500 to 900 °C the specific surface area dropped from around 531 to 285 m²/g, respectively. Nitrogen content decreased from around 10% down to 5%, respectively, while Raman spectra
recorded an increase in D-band to the G-band intensity ratio with the increase in carbonization temperature, which is an indication in increase in graphitization degree. Similar results, using pure chitin nanofibers, were reported in a separate study that showed as the carbonization temperature increased, from 500 to 900 °C, specific surface area decreased from around 458 to 263 m²/g, with increase in pore size from 2.75 to 2.86 nm (Hao et al. 2018a). The results also showed graphitization degree increases with the increase of carbonization temperature. Heteroatoms doping, oxygen and nitrogen doping, of CCNFs was synthesized from an insect, namely cicada slough (Zhou et al. 2017). The chitin powder obtained from cicada slough was mixed with physical activation agent, KOH at different weight ratios, (KOH:chitin) 1:2, 1:1, 2:1, and 3:1, before carbonization at 800 °C. Results showed that oxygen amount increased as KOH content increased, and vice versa for the nitrogen amount. Although at weight ratio of 3:1, results showed that CCNFs possess high surface area and total pore volume of 2217 m²/g and 1.02 cm³/g, respectively, and the carbon yield was at 10.9%. It was also shown that as the weight ratio increased the carbon yield decreased, while specific surface area increased. At weight ratio of 1:2 the surface area and total pore volume were shown to be 1243 m²/g and 0.57 cm³/g, respectively, and carbon yield was at 38.2%. These results shed the light on the possibility of synthesizing oxygen- and nitrogen-doped CCNFs and controlling the surface area and carbon yield. CCNFs offer finer nanofibers compared with most synthetic-based CNFs but lack the ability to form stand-free membranes due to the nature of chitin extraction conditions.

**Protein**

Protein is an abundant source for CNFs but lacks the ability to form a stand-alone CNF membrane. To tackle this issue, one group suggested the incorporation of lignin into plant protein to form CNFs. Though the structure of the membrane was maintained, the average fiber diameter increased drastically from 540 to 2610 nm after lignin addition (Yang et al. 2018a). Other proteins, such as prolamin, lose their fibril structure upon carbonization. Thus as a remedy solution, the addition of calcium salt, on one hand, enhanced the thermal stability and retained nanofibril structure, on the other hand, however, resulted in brittle CNFs (Yang et al. 2018b). To fabricate stable and flexible prolamin protein-based CNFs, the same previous group has suggested the utilization of transition metals, zinc, cobalt, and nickel (Wang et al. 2017b). They also followed a meticulous carbonization procedure after determining degradation temperature range of prolamin protein which was between 200 and 300 °C. First, the sample was heated to 200 °C at a rate of 5 °C/min and maintained for 2 h. Second, at lower heating rate of 1 °C/min the temperature was increased to 300 °C and maintained for 4 h. Finally, at heating rate of 1 °C/min the temperature was increased to 800 °C and maintained for 2 h before it was allowed to cool down to room temperature. The notable issues of protein-based CNFs are brittleness and free-standing capability that are yet to be addressed thoroughly.

**Others**

Biowaste-based nanofibrous and 1d-structures are rather limited compared with 0d-structure. For instance, carbonized kiwi fruits have shown to yield only partial carbonized nanofibrous structure along with nanosheet structure (Cheng et al. 2020). With a more distinct hollow nanofibrous structure, hexagonia apiaria, a type of fungus, has shown capability to preserve this distinct hollow nanofibrous structure after activation with KOH and carbonization at 800 °C for 2 h (Deng et al. 2017). The fiber diameter of hexagonia apiaria decreased from 2 µm to 620 nm after carbonization and was reported to have high specific surface area of 1280 m²/g. A group of researchers managed to synthesize magnetic nanofiborous carbon composite as sawdust as the carbon precursor (Liu et al. 2014). Briefly, purified sawdust waste, was mixed in Fe precursor, FeCl₃ solution, at elevated temperature, and the resultant dried material was carbonized. The nanofibers growth was thought to be as a combination of the high temperature and the role of catalyst played by Fe species mimicking chemical vapor deposition method. The specific surface area of the fibers was recorded to be around 360 m²/g.

The use of electrospinning to prepare nonconventional biomass and biowaste-based carbon nanofibers is rather limited because the process strongly depends on the spinnability of the biomass precursor, which is usually poor. Depending on the nature of the starting
| Carbon precursor | Materials processing conditions | Carbon conversion conditions | Carbon yield (%) | Ref |
|------------------|---------------------------------|-----------------------------|------------------|-----|
| Cellulose acetate | Electrospinning – Acetone and DMAc | Atmosphere@240 °C @3 °C/min for 1 h | – | Deng et al. (2013) |
| Cellulose acetate | Electrospinning – Acetone and DMAc | Atmosphere@240 °C @5 °C/min for 1 h | – | Li et al. (2019b) |
| Cellulose acetate | Electrospinning – Acetone and DMAc | Nitrogen @800 @5 °C/min for 2 h | 20 | Kuzmenko et al. (2014) |
| Cellulose acetate | Electrospinning – – – 0.1 M NaOH | Nitrogen @850 for 2 h | – | Cai et al. (2015) |
| Cellulose acetate | Electrospinning PAN DMF | Atmosphere@240 °C for 2 h | – | Li et al. (2019a) |
| Cellulose nanowhiskers | Self-assembly (Hydrolysis process) Microcrystalline cellulose | Centrifugation & freeze-drying | – | Cho et al. (2015) |
| Cellulose nanowhiskers | Self-assembly (Hydrolysis process) Cotton 65% H₂SO₄ | Centrifugation | – | Araujo et al. (2016) |
| Bacterial cellulose | Self-assembly (Gelation process) Nata-de-Coco Deionized water | Freeze-drying | – | Lee et al. (2013) |
| Bacterial cellulose | Self-assembly (Gelation process) BC pellicles – – Deionized water | Deionized water | 2.3 | Yu et al. (2014) |
| Bacterial cellulose | Self-assembly (Gelation process) BC pellicles – – Deionized water | Liquid nitrogen and freeze-drying | Nitrogen@850 @3 °C/min for 1 h | Jiang et al. (2016) |
| Bacterial cellulose | Self-assembly (Gelation process) BC pellicles – – Deionized water | Liquid nitrogen and freeze-drying | Argon@1300 for 6 h | Yang et al. (2019) |
material, spinnability simply could be improved by blending with an auxiliary polymer. For example, by employing electrospinning using a mixture of PEO and sodium alginate followed by a treatment in ionic cobalt solution, alginate nanofibers decorated with cobalt nanoparticles were fabricated (Li et al. 2015a). The electrospun nanofibers were carbonized in ammonia atmosphere before immersion in acid treatment. The resultant carbonized alginate nanofibers had distinct half-sphere mesopores between 10 to 40 nm and a diameter of around 100 nm as shown in Fig. 5. It was argued that during acid treatment, cobalt nanoparticles were removed leaving behind the mesopores, and that the ammonia environment was a key role to aid in graphitization process and forming desirable defects sites that enhance electrochemical properties. Another method features using electrospinning and liquefied walnut shell followed by carbonization to synthesize walnut shell-based nanofibers has been reported (Tao et al. 2017). The calculated average diameter was around 175 nm and had a specific surface area of around 408 m²/g. Since electrospinning offers great control on the morphology of the nanofibers, more efforts are needed to prepare tailored unconventional biomass and biowaste-based carbon nanofibers by means of developing spinnable biomass and biowaste materials.

To summarize, lignin, cellulose, chitin, and other biomass materials can be considered as the modern precursors for CNFs. Such materials not only provide us a renewable and sustainable option but also can be easily manipulated to obtain a tailored nanostructure with substantial physical properties suitable for energy applications. Table 5 summarizes the physical properties of different biomass- and biowaste-based CNFs.

It is worth to emphasize the different methodologies of KOH activation treatment due to their impact on the physical properties of carbon electrodes as is evident in Tables 2, 3, and 5, and throughout the reviews. The application of the KOH treatments differs depending on the adopted protocol. The most conventional method involves activation by soaking pre-carbonized materials into KOH solution or with a mixture of KOH and other activation agents. Solution concentration, mixture ratio, activation time, and activation temperature are the effective parameters that control the final physical properties of the carbon electrodes (He et al. 2016; Shehnaz et al. 2018; Zubbri et al. 2021). Others have utilized less common approach to active their samples using KOH as the activation agent. For example, one group utilized

![Fig. 5 a, b SEM and c, d TEM images of half-sphere mesopores alginate nanofibers carbonized at 600 °C. Reprinted with permission from Li et al. (2015a). Copyright (2015) American Chemical Society](image-url)
| Carbon precursor | Additive/activation agent | Doping conditions | \( I_D/I_G \) | \( d_{(002)} \) (nm) | Specific surface area (m²/g) | Total pore volume (cm³/g) | Pore size (nm) | References |
|------------------|---------------------------|------------------|--------------|----------------|------------------------------|------------------------|----------------|------------|
| Organosolv lignin | Graphene nanosheets/KOH | Self-doping | 12 – – 0.7 (S) | 0.88 0.34 | 2439 | 1.29 | 2–100 | Dai et al. (2019) |
| Alkali lignin     | MgO–                      | None             | – – – – – | 0.98 – | 1140 | 0.627 | 2–100 | Ma et al. (2018) |
| Alkali lignin     | Mixture of CO₂ and N₂ | None            | – – – – | 0.98 – | 2170 | 0.987 | 2–4 | Jayawickramage and Ferraris (2019) |
| Low sulfonate alkali lignin | NaOH | None             | – – – – | 0.91 – | 1444 | 0.91 | 0.7–1.5 | Hu and Hsieh (2013) |
| Bacterial cellulose | Potassium citrate/– | None            | – – | 0.89 – | 1037 | 1.04 | 3.8–9.6 | Jiang et al. (2016) |
| Bacterial cellulose | –/– | Self-doping | 2.3 – | 1.9 0.38 | 128 | – | – | Yang et al. (2019) |
| Chitin            | –/KOH                     | Self-doping | 1.5 33.8 – | – – | 2217 | 1.09 | 1.5 | Zhou et al. (2017) |
| Chitin            | –/–                       | Self-doping | 7.3 6.6 – | 1.03 0.39 | 369 | – | 4.3 | Hao et al. (2018b) |
| Protein(hordein &zein)-lignin | –/CO₂ | Self-doping | 1.3 4.3 – | 1.01 0.38 | 1113 | – | 1.4 | Yang et al. (2018a) |
| Plant protein     | Ca(OH)₂/–                 | Self-doping | 6.4 7.7 – | 1.02 0.37 | 348 | – | 1.9–8.7 | Yang et al. (2018b) |
| Fungus (hexagonia apiaria) | –/KOH | Self-doping | 1.9 5 – | 1.55 – | 1280 | – | 2.2 | Deng et al. (2017) |
ultrasonic treatment to impregnate KOH on CNFs (Dai et al. 2019). Recent KOH activation methods involve assisted tool to minimize cost and time have also been reported. For instance, a group demonstrated that KOH treatment polysized in a microwave reactor can be effectively done within short time, 10 min (Feng et al. 2020). While another group showed that laser writing is also an effective method for KOH activation treatment since it’s cost and time effective method (Liu et al. 2020b).

Most biomass and bio-based carbon precursors contain different chemical groups, thus making the KOH activation treatment differs from the activation treatment in carbon materials of petroleum nature. A recent study attempted to explore the role of KOH activation in biomass rich with oxygen groups (Chen et al. 2020b). They utilized Bamboo waste as their starting materials. As the result of abundant oxygen species, they found that activation with KOH can be effectively done at low impregnation ratios and low temperatures. Furthermore, activation can be further improved by allowing KOH to further react with carbon at slightly higher temperatures or higher impregnation ratios. It was argued that at low temperatures KOH reacted with the biomass and completely converted into K$_2$CO$_3$ accompanied with the release of byproducts such as phenols. As the temperature increased, K$_2$CO$_3$ was transformed to K$_2$O, and phenols release dropped. At the end of the activation process, stable oxygen groups were present and stable.

2D-nanostructure

This category is composed of bio-based carbonized thin films or papers with distinct nanocrystalline, and/or nanoporous structures.

Lignin

The potential of lignin-based carbon thin films in energy applications is still relatively new. The first characterization of a lignin-based carbon thin film is dated back to 2007 (Shen and Zhong 2007). The pore size and porosity of the film were found to be influenced by the amount of lignin (8–20 wt.%%) that in turn influenced the UV adsorption and electric resistance properties of lignin-based carbon thin film. Recently, more complex lignin-based carbon thin film structures have been developed (Sun et al. 2019a; Luo et al. 2019). Interestingly, the addition of NiCl$_2$ as a catalyst and Na$_2$CO$_3$ as both a template and an activator to lignin solution had a pronounced effect on the physical properties of lignin-based carbon thin film (Sun et al. 2019a). Specific surface area of pristine lignin-based carbon thin film carbonized at 1000 °C was measured to be around 287 m$^2$g$^{-1}$, while lignin-based carbon thin film incorporated with Na$_2$CO$_3$ and treated with NiCl$_2$ carbonized at the same temperature was measured to be around 730 m$^2$g$^{-1}$. This increase of surface area was attributed to pores generated by the formation of Ni nanoparticles that helped form pores and channels, and to the decomposition of Na$_2$CO$_3$ into gases at different stages during carbonization that helped form more pores. Investigation of new activators and lignin’s solvents, may prove vital for the development of lignin-based carbon thin films and the enhancement of their physical properties.

Cellulose

Pristine cellulotic-based and hierarchical composite nanostructured carbon films have attracted the attention of many research groups (Vuorema et al. 2010; Li et al. 2017a; Hwang et al. 2018; Meng et al. 2019). At a low-cost, a high degree of graphitization of pristine CNC film was fabricated at a carbonization temperature of 1000 °C (Zhu et al. 2017). Results illustrated surface area and pore sizes of 146 m$^2$g$^{-1}$ and 1.7–30 nm, respectively. A simple and straightforward strategy was developed to prepare flexible film cellulose-based carbon nanoﬁbers ﬁlled with activated carbon (Li et al. 2016c). First, cellulose and activated carbon were thoroughly mixed together and vacuum-dried, was carbonized at 800 °C for 2 h. The carbonized film preserved its integrity perhaps due to the available functional groups on cellulose nanoﬁbers, e.g. hydroxyl group, that enhanced the interconnection between the two materials. The morphology of the film is depicted in Fig. 6. Results showed that the carbonized film had a large surface area of 1840 m$^2$g$^{-1}$ which was due synergetic effect of the readily large surface area of the activated carbon and carbonized cellulose nanoﬁbers. In a separate study, a combination of cellulose nanofibrils and cellulose nanocrystals (CNC) was prepared as nanostructured carbon film (Li et al. 2017b). But in order to avoid aggregation
between cellulose nanofibrils and CNCs, a sacrificial 5 nm thick layer of Al$_2$O$_3$ was deposited using atomic layer deposition on the uncarbonized cellulose nanofibrils/CNC film. The layer of Al$_2$O$_3$ was able to penetrate inside the sample through mesopores, thus, avoiding aggregation during carbonization which was carried out at 900 °C. The partially graphitized nanostructured carbonized film not only maintained nanofibrous and nanoporous structures but also exhibited large surface area, large total pore volume, and small pore size of around 1244 m$^2$/g, 2.2 cm$^3$/g, and 3.5 nm, respectively. Effort to fabricate a 2D-nanostructured composite film is noted too. A flexible 10 μm thick composite composed of carbonized cellulose paper and graphene/thin-graphite layer was fabricated in a one-step process using microwave plasma enhanced chemical vapor deposition (Ren et al. 2016). Edge oriented graphene sheets were grown on the paper, which was simultaneously carbonized during the process. The graphene growth and cellulose carbonization process were done in only 5 min. The carbon fibers are hollow filled with micro- and mesopores on their surfaces.

**Chitin**

Carbon nanoporous films have also been prepared from chitin derivatives. Using silica as a sacrificial material, chitin-based carbon film with high surface area and large pore volume was prepared (Nguyen et al. 2014). At the optimum silica concentration, surface area and pore volume were reported to be 1130 m$^2$/g and 1.0 cm$^3$/g, respectively, which were attributed to the space that was created by silica matrix during carbonization. In addition, silica content might have increased carbon yield too. Unique and scalable composite honeycomb-like chitin and reduced graphene film was also prepared (Wang et al. 2016). Before carbonization, the process consists of freezing and thawing cycles of a dispersion of chitin

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**Fig. 6** SEM images at different magnifications of cellulose nanofibrils- and cellulose nanocrystals-based carbon film A, B before carbonization, C–E after carbonization, and F TEM image of single cellulosic carbon fiber. Reprinted with permission from Li et al. (2016c). Copyright (2016) Elsevier
and graphene oxide in NaOH-urea-water solution followed by coagulation using ethanol. Large pore interconnected structure, 1–3 μm, which is the result of phase separation, was attributed to the formation of honeycomb structure.

Others

There have been very few recent reports utilizing unconventional biomass and biowaste materials such as glucose, rice straw, and poly(furfuryl alcohol) as carbon precursors to develop nanostructured films (Hawes et al. 2019). The reported films were prepared by filtration method. Using infrared (IR) laser writing as the carbonization tool, carbonized poly(furfuryl alcohol) (PFA), a polymer that was derived from a biowaste, and reduced graphene composite (rGO) composite was prepared (Hawes et al. 2019). It was reported that the carbonization of PFA film using IR laser was not possible unless the medium contains some loads of graphene oxide (GO). In the composite, the laser intrigued the carbonization of PFA film and the reduction of GO to rGO sheets. The resultant morphology of the film was composed of PFA-based carbonized spheres with an average diameter of 670 nm with distributed rGO sheets that have lateral size ranging from 100 to 1400 nm. Another approach to prepare 2D-nanostructured carbon composite film using cotton fabrics as a substrate was also noted (Li et al. 2020). Nonetheless, binder-free 2D-nanostructured films and papers derived from biomass and biowaste materials are limited.

3D-nanostructure

This category is composed of bio-based 3D interconnected networks with distinct nanostructure features such as aerogel and foams.

Lignin

Aerogels are of low density and porous 3d-nanostructures. The preparation of lignin-based aerogels has been investigated by many groups. The main material parameters that go into the formation of lignin aerogels are (i) content of lignin and its molecular weight, (ii) auxiliary resins and their mass ratios, (iii) catalyst content, and (iv) extraction conditions, e.g. freeze-drying or CO2 supercritical drying (Chen and Li 2010; Chen et al. 2011; Grishechko et al. 2013a). The initial amount of lignin and its molecular weight influence both gelation time and pore structure of the carbonized aerogel (Seo et al. 2014; Yang et al. 2017). The most notable auxiliary resin mixtures that have been studied are phenol–formaldehyde (Grishechko et al. 2013a), resorcinol–formaldehyde (Chen and Li 2010; Chen et al. 2011), and tannin–formaldehyde (Grishechko et al. 2013b), and NaOH and Na2CO3 are the most widely used catalysts among many others. These parameters have a pronounced effect on the final properties. For example, an interesting report showed that copper-doped lignin carbon aerogels resulted in higher porosity and specific surface area, 67% and 431 m²g⁻¹, respectively, compared with pristine lignin-based carbon aerogel, 55% and 162 m²g⁻¹, respectively (Xu et al. 2018a). In this example, copper played the role of the catalyst during sol–gel process, and the role of activator during carbonization. Table 6 summarizes some of these parameters and their corresponding physical properties for lignin-based and other biomass carbon aerogels.

The main advantage of lignin is that it has reactive sites, e.g. para carbon and ortho meta, that are available to react with formaldehyde as crosslinker, which lower the need to crosslinking in the presence of phenol or resorcinol (Grishechko et al. 2013a; Xu et al. 2015), unfriendly substances. Worth reporting that lignin-based carbon aerogels have shown to be fabricated without using phenol or resorcinol (Yang et al. 2017). Nonetheless, the main disadvantage of lignin-based aerogels is that they are brittle in nature which limits applications scope. To tackle brittleness of lignin aerogels, in one study, bacterial cellulose was incorporated to lignin to increase the elasticity of lignin-based carbon aerogels at a reversible strain of 20%, albeit with a low reported compressive strength of only 0.03 MPa (Xu et al. 2015).

Cellulose

Unlike lignin-based carbon aerogels, which typically involve resorcinol–formaldehyde as their main constituent, most of cellulose precursors, i.e. cellulose nanofibers, cellulose microcrystalline, and bacterial cellulose, could be used without auxiliary chemicals to fabricate carbon aerogels.

First, cellulose nanofibers have been used to form aerogels. For example, nitrogen doped cellulose-
| Carbon precursor          | Solvent/concentration (wt.%) | Catalyst       | Auxiliary resin | Additive                          | Gel time temperature (°C)/time (hours) | Drying method            | Density (mg cm⁻³) | Porosity (%) | SSA* (m²/g) | Total pore volume (cm³/g) | Pore size (nm) | References                |
|--------------------------|------------------------------|----------------|-----------------|-----------------------------------|----------------------------------------|--------------------------|------------------|--------------|-------------|-----------------------------|-----------------|--------------------------|
| Organosolv lignin        | Water/20                    | NaOH           | Formaldehyde    | –                                 | 85/2                                   | Freeze-drying            | 0.005–1.0        | > 0.45       | 19          | 0.007                       | 1.5             | Yang et al. (2017)        |
| Kraft lignin             | Water/20                    | NaOH           | Formaldehyde    | –                                 | 85/16                                  | Freeze-drying            | 0.005–1.0        | > 0.45       | 121         | 0.45                        | 15              | Yang et al. (2017)        |
| Alkali lignin            | Water/17                    | NaOH & Na₂CO₃  | Resorcinol/formaldehyde | Bacterial cellulose                | –                                      | CO₂ supercritical liquid | 2.6              | –            | 190         | –                           | 17.8            | Xu et al. (2015)          |
| Enzymatic hydrolysis lignin | Water/–                     | Na₂CO₃         | Resorcinol/formaldehyde | –                                 | 80/9                                   | Ambient drying in acetic acid | –                | 60.4         | 779         | 0.48                        | 9.4             | Xu et al. (2018b)         |
| Alkali lignin            | Water/–                     | –              | Resorcinol/formaldehyde | CuCl₂·6H₂O                         | –/0.17                                 | Freeze-drying            | –                | 54.5         | 162         | 0.11                        | –               | Xu et al. (2018a)         |
| Alkali lignin            | Water/–                     | –              | Resorcinol/formaldehyde | CuCl₂·6H₂O                         | –/0.17                                 | Freeze-drying            | –                | 47.6         | 899         | 0.63                        | –               | Xu et al. (2018a)         |
| Fe₂O₃-bacterial cellulose | –                            | –              | –               | –                                 | –                                      | Freeze-drying            | –                | –            | 327         | 0.59                        | –               | Huang et al. (2016)       |
| N-doped bacterial cellulose | –                            | NH₃            | –               | –                                 | –                                      | Freeze-drying            | –                | –            | 875         | 0.78                        | 3.0             | Zhu et al. (2018a)        |
| Chitin                   | phytic acid & hydrogen/8 & 3 | –              | –               | –                                 | 110/6                                  | Freeze-drying            | –                | –            | 586         | –                          | < 2             | Gao et al. (2019)         |
| Shrimp shell chitin      | NaOH-urea-H₂O/11:4:85 wt.%  | –              | –               | –                                 | 50/2                                   | Freeze-drying            | –                | –            | 521         | 0.3                         | < 2             | Dassanayake et al. (2018) |
| Partially deacetylated chitin | Water/0.5—2.5               | NaOH           | –               | NH₃ & EtOH                         | –                                      | Freeze-drying            | –                | –            | 1597        | 3.2                         | 4.1             | Ding et al. (2018)        |
| Soy protein              | Water/–                     | –              | Cellulose       | –                                 | 180/5.5                                | Freeze-drying            | –                | –            | 697         | 0.38                        | ~ 5             | Alatalo et al. (2016)     |
| Agaric                   | –                            | NH₄Cl          | –               | –                                 | –                                      | Freeze-drying            | –                | –            | 2200        | –                          | 5–10            | Zhang et al. (2019a)      |
| Konjac glucomannan       | Water                       | NaOH           | SiO₂ nanofibers | –                                 | –                                      | Freeze-drying            | 0.14              | 99.99        | –           | –                          | –               | Si et al. (2016)          |
| Durian                   | –                            | –              | –               | –                                 | –                                      | Freeze-drying            | 618               | 0.43         | > 5         | –                          | –               | Lee et al. (2020)         |
| Jackfruit                | –                            | –              | –               | –                                 | –                                      | Freeze-drying            | 511               | 0.28         | > 5         | –                          | –               | Lee et al. (2020)         |

*SSA refers to specific surface area
nanofibers aerogels reinforced with GO was fabricated and carbonized to convert the cellulose-nanofibers to carbon and the graphene oxide to reduced graphene resulting in cellulose nanofibers-based reinforced rGO carbon aerogel (Zhang et al. 2018). Briefly, the nitrogen doping was possible by preparing a homogeneous mixture of melamine, an organic material rich with nitrogen, and cellulose nanofibers in one pot. The resulted mixture was mixed with a predetermined amount of GO solution. The composite mixture was allowed to be dried in nitrogen liquid before initiating freeze-drying followed by carbonization at 600 °C. The carbonized aerogel resulted in a specific surface area of 487 m²/g, while its rich nitrogen content facilitated hydrophilicity, and the rGO allowed the aerogel to exhibit 40% recoverable volume change under compression loading. Another example illustrates hierarchical structure of cellulose-nanofibers composite aerogels decorated with NiCo₂S₄ nanocrystals (Liu et al. 2020a). The cellulose-nanofibers were freeze-dried to form aerogel then immersed in NiCo₂S₄ S₄ precursor before initiation of a hydrothermal reaction and carbonization at low temperature of 500 °C. It was found that the carbon aerogel composite has a relatively high specific surface area and large pore volume of 394 m²/g and 0.9 cm³/g, respectively.

Secondly, a recent study showed that another source of cellulose, namely cellulose microcrystalline, dried in a supercritical CO₂ was possible to produce carbon aerogels with large surface area and pore volume, 892 m²/g and 1.80 cm³/g respectively, which substantially further increased after CO₂ activation, 1873 m²/g and 2.65 cm³/g respectively (Zu et al. 2016). Moreover, by comparing specific surface area and total pore volume of the nonactivated carbonized (carbonized at the same temperature 800 °C) and freeze-dried carbon aerogel, 418 m²/g and 0.75 cm³/g respectively, with their supercritical dried counterparts, 892 m²/g and 1.80 cm³/g respectively, it seems that supercritical drying might be more effective. This was attributed to the inner structure difference where supercritically dried carbon aerogels are characterized as homogenous and its nanofibrous structure tending to aggregate less. Worth reporting that the process of making based carbon aerogels from cellulose microcrystalline is more complex than previous cellulosic nanofibers because it involves preparation of nanocellulose gels and regeneration before obtaining aerogel structure.

Thirdly, BCs have been carbonized to prepare carbon aerogels. The preparation of BC as carbon aerogel is a two-step process: freeze-drying of BC aerogels, to preserve nanofibril structure, and carbonization at high temperature to convert BC aerogels to low dense BC-based carbon aerogels (Wu et al. 2013; Huang et al. 2015). Other practices may involve freezing at -196 °C in liquid nitrogen prior to freeze-drying (Liang et al. 2012; Wang et al. 2014, 2020a) or freezing in supercritical CO₂ (Liebner et al. 2010). BC-based aerogels, filled with 3D nanofibrous structure with a diameter equal or less than 20 nm, are flexible and tend to be less dense, 0.004 to 0.008 cm³/g (Liebner et al. 2010; Wu et al. 2013), than their traditional carbon counterparts, 0.15 to 0.75 cm³/g (Fu et al. 2003; Wu et al. 2004). Depending on the initial size of BC pellicles and the fact that around 15% of the initial volume will be lost, carbonized BC aerogels size and volume can be tuned. In addition, BC aerogels can afford reduction in volume up to 90% when the carbon aerogels are under compression loads and return to their original shape after removal of loads (Wu et al. 2013). BC-based carbon aerogel hierarchical composites have also been developed (Wan et al. 2015; Huang et al. 2016; Zhuo et al. 2019; Wang et al. 2019a). For example, BC-based carbon aerogel decorated with iron oxide nanoparticles was developed with a specific surface area of 322 m²/g (Wan et al. 2015). Briefly, after obtaining BC aerogels in liquid nitrogen, the as-prepared aerogels immersed in Fe(NO₃)₃,9H₂O solution for subsequent hydrothermal synthesis to obtain Fe₂O₃ nanoparticles. The resulted composite was freeze-dried and carbonized at 600 °C to obtain BC-based carbon aerogels decorated with Fe₂O₃ nanoparticles.

BC-based nitrogen-doped carbon aerogels can be prepared simply by thermally treating BC pellicles in ammonia environment followed by carbonization as illustrated in Fig. 7. Such treatment further enhances physical properties. For example, specific surface area and pore volume of BC-based nitrogen-doped carbon aerogels were found to be 875 m²/g and 0.78 cm³/g, respectively. While specific surface area and pore volume of pristine BC-based carbon aerogel were 585 m²/g and 0.55 cm³/g, respectively. Plus, nitrogen doping decreased graphitization degree by imparting defects (Zhu et al. 2018a). Nitrogen-doping has also been featured for BC-based carbon aerogels. Recent studies have demonstrated that nitrogen-doping in BC
could be obtained by incorporating the 3D structure with nitrogen-rich nanocrystals such as zeolitic imidazolate frameworks, e.g. ZIF-8 and ZIF-67, that contain 2-methylimidazole which is rich in nitrogen species (Fei et al. 2020; Chen et al. 2019). For example, using ZIF-8 nanocrystals, it was shown that the nitrogen content could reach up to 7% at a carbonization temperature of 900 °C (Chen et al. 2019).

Rapid microwave plasma pyrolysis as an alternative carbonization method has been reported to carbonize BC (Islam et al. 2017). The procedure was done in microwave plasma enhanced chemical vapor deposition system in methane and hydrogen environment under controlled pressure. The estimated temperature of carbonization was around 1200 °C. The entire plasma pyrolysis had lasted only for 15 min, and the pyrolyzed BC aerogel mimicked its nanofibrous structure. The reported surface area, 57.5 m$^2$/g, however, is inferior to traditionally carbonized BC aerogels. Overall, cellulosic carbon aerogels prepared from BC not only are easy to prepare compared with other cellulosic precursors, but also show high quality porous inner nanostructure.

**Chitin**

Depending on chitin source and preparation protocols, chitin-based carbon aerogels have successfully been developed with distinct inner structures. Usually, chitin aerogels have nanofibrous inner structure with a fine diameter of 10 nm (Nogi et al. 2010). Unlike cellulose- and lignin-based carbon aerogels, chitin-based carbon aerogels already contain high amount of nitrogen atoms depending on carbonization conditions, for example, at 600 and 900 °C the nitrogen content is around 8% and 6%, respectively (Nogi et al. 2010; Li et al. 2015c). Moreover, self-doping of heteroatoms, nitrogen and oxygen atoms, is possible too. For example, nitrogen- and oxygen-rich chitin-based nanosheets carbon aerogels with high surface area of 586 m$^2$/g and average pore size less than 2 nm have been developed (Gao et al. 2019). The protocol starts by dispersing chitin powder in phytic acid and hydrogen peroxide solution before placing the mixture in autoclave environment at elevated temperature of 110 °C for 6 h. Subsequently, after rinsing with water, the mixture was freeze-dried before carbonization. The attributed large surface area was thought to be related to the decomposition of nitrogen and oxygen content during carbonization.

Depending on the starting chitin, different preparation methods have been reported. One approach involves the use of sol–gel method (Ding et al. 2012; Dassanayake et al. 2018). First, chitin powder was dispersed in NaOH-urea-water solution at low temperature, −20 °C, before thawing (Dassanayake et al. 2018). Freezing and thawing cycles were repeated more than once for the solution before subjecting the obtained transparent solution to gelation process at elevated temperature, 50 °C. After washing with water, the hydrogel was freeze-dried at −105 °C. The chitin aerogel was carbonized at 800 °C and resulted in high specific surface area. Another different approach illustrated in Fig. 8 involves using deacetylated chitin nanofibers dispersed in NaOH solution after weak acid treatment (Ding et al. 2018). Next, a hydrogel process was initiated by ammonia and ethanol gas coagulation to form hydrogen cross-linking at room temperature. Before freeze-drying, solvent exchange was carried out using t-BuOH. The nanofibrous chitin aerogel was carbonized at 500, 700, 900, and 1000 °C for 2 h. The nitrogen and oxygen content of chitin-based carbon...
aerogel decreased as the carbonization temperature increased. However, when the carbonization temperature increased from 500 to 1000 °C, the surface area and pore volume substantially increased from 528 to 1597 m²/g and from 1.5 to 3.2 cm³/g respectively, while average pore size decreased from 5.7 to 4.1 nm, respectively. A similar decreasing trend was observed for graphitization degree as the carbonization temperature increased, Raman intensity ratio of $I_D/I_G$ (intensity ratio of D band to G band) decreased from 1.02 to 0.89, respectively.

Hierarchal chitin-based carbon complex composites are another possible structure. For instance, nanohybrid chitin/cellulose/NiFe$_2$O$_4$ composite aerogel rich with nitrogen was synthesized (Liu et al. 2020c). Briefly, a homogenous composite dispersion of chitin/cellulose/NiFe$_2$O$_4$ synthesized in the presence of NaOH was heat treated in autoclave environment at 180 °C to form hydrogel once cooled down. After thoroughly washing with water, the chitin/cellulose/NiFe$_2$O$_4$ composite hydrogel was freeze-dried to obtain aerogel. Finally, at different temperatures the composite aerogel was carbonized. Before freeze-drying, the nanofibers composed of both chitin, 8 nm in diameter, and cellulose, 3–4 nm in diameter, was thought to be used as nucleation sites to homogenously growing NiFe$_2$O$_4$ nanoparticles with a diameter of 10 nm. However, after carbonization and freeze-drying, the nanofibrous structure along the nanoparticles became nanofibrous/nanosheets aerogel rich with nitrogen atoms. In addition, the size of nanoparticles increased dramatically to around 100 nm, which could be controlled by controlling the carbonization temperature and duration.

**Protein**

Protein-based carbon aerogels are rich with heteroatoms, especially nitrogen atoms, however, are brittle in nature. The brittleness has shown to be overcome by adopting protein-based composite instead. Such composites are made of protein sources, e.g. ovalbumin, soya bean flour, or silk regenerated proteins, and carbohydrates, e.g. cellulose and glucose, or graphene oxide (White et al. 2011; Yun et al. 2014a; Alatalo et al. 2016). For instance, a composite carbon aerogel was prepared from silk regenerated proteins, extracted from cocoons, and graphene oxide (Yun et al. 2014a). The composite was carbonized at 800 °C for 2 h to convert proteins to carbon and graphene oxide to rGO. Although the composite aerogel has a pore size of around 11 nm, the specific surface area, 181 m²/g, is very modest compared with other carbon aerogels. Another example, soy bean flour, as the protein source, mixed with either glucose or cellulose were carbonized at 1000 °C to fabricate carbon aerogels.
(Alatalo et al. 2016). Results showed that soy protein-glucose-based carbon aerogel has acceptable specific surface area of around 449 m²/g and total pore volume 0.25 cm³/g, while soy protein-cellulose-based carbon aerogel possesses higher specific surface area and larger total pore volume, 697 m²/g and 0.38 cm³/g, respectively, albeit its nitrogen content is lower. The difference in physical properties was attributed to the difference in the end physical structure of the composites, where glucose-based carbon aerogel resulted in aggregated nanoparticles and cellulose-based carbon aerogel resulted in fibril structure.

Others

Unconventional biowastes and biomass materials have been employed as carbon precursors to fabricate aerogels. Hydrothermal treatment accompanied with auxiliary chemical process have shown to be simple and rapid approach to fabricate biomass-based, e.g. glucose and agaric, hierarchical nanostructure carbon aerogels (Fellinger et al. 2012; Zhang et al. 2019a). For example, a hydrothermal treatment to agaric, type of mushroom fungus, solution followed by freeze-drying and carbonization chemical blowing resulted in agaric-based nanosheets carbon aerogel with high the specific surface area of 2200 m²/g (Zhang et al. 2019a). It was argued that the blowing gas, NH₄CL, decomposed into NH₃ and HCl gases that generate mesopores in the range 5 to 10 nm and also facilitated the formation of 1.5 nm thick nanosheets carbon layers. Another recent report has utilized hydrothermally treated aloe juice, a plant source, as carbon precursor to fabricate Co₃O₄/C composite hierarchical aerogel structure with high specific surface area, 728 m²/g (Yin et al. 2019). Figure 9 shows preparation step of aloe-based carbon composite and its surface morphology after freeze-drying, hydrothermal treatment, and carbonization. The corresponding Raman spectra and X-ray diffraction are also shown.

Unconventional carbon precursors that have been used as a starting material for aerogels include polysaccharides, waste foods, e.g. vegetables, fruits, and nuts, and biowastes, e.g. leather. Polysaccharide derivatives aerogels have also successfully been utilized as carbon precursors to form hierarchical aerogel structures (El-Naggar et al. 2020; Si et al. 2016). To illustrate, resilient konjac glucomannan-based nanofibrous carbon aerogel has been designed and fabricated (Si et al. 2016). SiO₂ nanofibers have been employed as a template to support the inner honeycomb structure of the aerogel. The resultant aerogel was reported to be highly elastic with ultralarge porosity of 99.99% and ultralow density of only 0.14 mg/cm³.

Waste foods and plant parts are excellent natural precursors for cheap and high-quality carbon aerogels. Many researchers have been investigating a wide range of biowaste organic foods, e.g. vegetables, fruits, and nuts, to fabricate high-performance electrodes in a form of 3D-structure. Cabbage waste has shown to be converted into carbon aerogels after a series of treatment, cryofreezing, freeze-drying, and carbonization (Cai et al. 2018). The partially nitrogen- and oxygen doped cabbage-based carbon aerogel have reported a high specific surface area of 536 m²/g and an average pore size of 1.9 nm. Employing a simple approach of hydrothermal treatment, freeze-drying and carbonization, a recent study has utilized the core of two tropical fruits, namely durian and jackfruit, to fabricate carbon aerogels (Lee et al. 2020). Durian- and jackfruit-based carbon aerogels exhibited specific surface area and pore volume of around 618 m²/g and 0.4 cm³/g, and 511 m²/g and 0.28 cm³/g, respectively. Both carbon-based aerogels showed relatively high heteroatoms doping that were considered as important pseudo-capacitance performance boosters. Likewise, KOH activated hydrothermally treated pear, a fruit, has been used to fabricate highly graphitized carbon aerogel with ultrahigh specific surface area 2323 m²/g (Myung et al. 2019). Other recent waste foods and plant parts as carbon precursors are potatoes and elm seeds (Lu et al. 2019; Guo et al. 2019).

Biowaste materials have engaged researchers’ attention to synthesize biowaste-based hierarchical carbon aerogels as high-performance electrodes for energy applications. For instance, leather, a natural material, wastes were treated with KOH solution before initiating freeze-drying and carbonization processes (Liu et al. 2020d). The low-dense lather-based aerogel, 0.56 mg/cm³, showed high specific surface area of 2523 m²/g and heteroatoms doping. Another example, sodium lignosulphonate, a biomass substance used in food industry, as carbon precursor and mesoporous silica template as a supporting template have been used to fabricate hierarchical ordered activated with ZnCl₂ (Bai et al. 2020). The aerogel had a large specific surface area and pore volume of 1481 m²/g and 2.62 cm³/g, respectively. Likewise,
wheat straw, harvesting biowaste, has been treated to fabricate lignocellulose-based 30 nm thick nanosheets carbon foam with large specific surface area and large mean pore size, 1063 m$^2$/g and 420 nm, respectively (Gou et al. 2020).

Bio-based nanostructured carbon materials for high-performance energy applications

Biomass and biowaste carbon precursors are abundant, environment friendly, and renewable materials. Such biomass and biowaste materials include lignin, cellulose, chitin, protein, and other unconventional biomass choices, e.g. food waste, flowers, seeds, plant
by-product wastes, etc. After carbonization, each of these materials possesses unique physical, chemical, and morphological properties qualifying it to be a high-performance electrode in energy storage devices, namely lithium-ion batteries and supercapacitors (Selvan et al. 2018; Li et al. 2018a; Zhu et al. 2020a).

The paradigm shift we are witnessing toward electric vehicles and high-performance electronic portables pushes the limit of energy storage devices every day. To meet such technological demand, new material preparation protocols have been researched and developed to reach high electrochemical performance in energy storage devices using carbon-based electrodes. Such protocols involve doping of single atoms or heteroatoms, increasing specific surface area, and enhancing physical structure. In the following sections, a review of the factors influencing electrochemical performance and summary of the most recent electrochemical performance of bio-based carbon electrodes in rechargeable batteries and SCs are given.

Factors influencing electrochemical performance of carbon electrodes

In energy storage devices, it has been established that N-doped (nitrogen-doped) carbon electrodes show better electrochemical performance than the same non-doped electrodes. For example, in SCs, N-doped carbon electrodes yielded lower resistance (0.23 $\Omega$) and higher capacitance (313 F g$^{-1}$ at 1 A g$^{-1}$) compared with non-doped carbon electrodes that yielded higher resistance (1.32 $\Omega$) and lower capacitance ($\sim$ 225 F g$^{-1}$ at 1 A g$^{-1}$) (Zhao et al. 2015a). It was also demonstrated that N-doped carbon electrodes are of hydrophilic nature (Zhai et al. 2011; Yang and Zhou 2017; Chen et al. 2017) while non-doped counterpart is of hydrophilic nature (Zhao et al. 2015a). Therefore, the enhancement in SCs using N-doped electrodes is attributed to the increased ion diffusion, improved wettability, and pseudocapacitance activity which is the result of nitrogen doping (Zhao et al. 2015a; Hou et al. 2015). Similar enhancement is seen in LIBs after nitrogen doping, which is attributed to the ability of lithium ions to be tucked and stored between the defect sites created by the presence of nitrogen functional groups (Zhang et al. 2014). Generally, N-doping can be realized by carbonizing enriched-nitrogen carbon precursors, e.g. PAN, at low temperature to maintain some of the original nitrogen content. Another important class of doping is sulfur-doping. Sulfur-doping enhances cycling stability, increases conductivity, and paves the way for more ions to be stored by providing extra reactive spots (Yun et al. 2014b; Li et al. 2015b). Additionally, oxygen- and heteroatom-doping have also shown to enhance the electrochemical performance of energy storage devices by increasing wettability, microporosity, and pseudocapacitance activity (Wu et al. 2012; Chen et al. 2014; Xu et al. 2017). For example, three-dimensional heteroatom-doping using phosphor, boron, and nitrogen atoms was shown to be an effective approach to improve electrochemical performance in SCs (Chen et al. 2014).

Another effective approach to enhance reach high electrochemical performance is by adopting materials with high specific surface area. The main virtues of high specific surface area in an electrode in energy applications are the extra space provided for ions in the electrolyte solution to be stored at higher rate, facilitation of ions diffusion, and lower electrical resistance (Yang et al. 2011; Liu et al. 2017b). It has been shown that electrodes with high specific surface area result in high-performance electrodes regardless of the initial starting materials, e.g. graphene or PAN (Zhang and Lou 2013; Zhang et al. 2013; Heo et al. 2019). Generally, for carbon-based electrodes, high specific surface area can be achieved by either physical or chemical activation or by incorporation of nanoparticles to increase roughness of the surface as is discussed in the previous section.

Physical structure is mainly related to the stability of the electrode which provides a better medium to store energy. That could be controlled by modifying the preparation protocol to involve steps promoting stable physical structure, for example, freeze-dried carbon-based aerogels allow for a stable physical structure and a control over the nanoporous structure that results in electrodes of good electrochemical stability and high-performance (Jung et al. 2015; Zhou et al. 2018). The physical structure of carbon materials could be further enhanced by allowing graphene-like nanosheets to be apart from each other so that an extra space is available for ions to be tucked in and stored. This approach was best illustrated in a recent work particularly for supercapacitors (Dai et al. 2019). It was shown that the additional spaces provided by
graphene-like nanosheets not only improved electrochemical performance but may be also was advantageous to the cycle life of the device.

High-performance bio-based nanostructured electrodes in rechargeable batteries

Efforts to utilize carbon-based materials in energy storage devices as functional electrodes have been widely noted among many research groups due to their chemical and electrochemical stability and charge storage ability (Zhu et al. 2015, 2016, 2020a; Li et al. 2018b). Conventional batteries consist of electrodes, electrolyte, and a separator. In principle, lithium ions flow in the electrolyte (e.g. lithium hexafluorophosphate dissolved in DMC or EC. See Table 7 for more details) from cathode and intercalates on anode during charging, and flow in reverse during discharging. The anode and cathode are physically separated by a porous separator to prevent short circuit control flow of ions.

Five of the most researched batteries are lithium-ion battery (LIB), lithium-oxygen battery (Li–O), lithium-sulfur battery (Li–S), sodium-ion battery (NIB), and potassium-ion battery (KIB) due to their abundance in Earth and good performance (Ji 2019). The advance of their electrochemical performance is mainly tight with the advance of the electrodes by extending the life cycle and increasing the reversible capacity. In batteries, efforts aimed at fabricating high-performance electrodes to meet the ever-increasing technological demands. For many groups, developing and testing the performance of carbon-based electrodes in batteries have been their immediate aim (Zhu et al. 2016, 2018b; Li et al. 2018a).

The most recent research has been focusing on utilizing biomass or biowaste materials that delivers high degree of oxygen and nitrogen species to allow for self-doing mechanisms after carbonization, or that delivers high specific surface area, or a combination of the two (Gao et al. 2017b). They also focused on artificially sulfur-doping the initial starting materials to allow for heteroatoms- or co-doping in order to enhance the overall performance of the battery. To demonstrate, as shown in Fig. 10, in LIBs and Li-Ss, large reversible specific capacities exceeding 500 mAhg\(^{-1}\) limit with electrochemical stability over 100 cycles have been demonstrated by utilizing honeycomb-like carbon electrode derived from bagasse (Wan and Hu 2020). Such high-performance was mainly attributed to the synergetic effect of the sulfur and oxygen species hanging from the honeycomb-like structure. For both LIBs and Li-Ss, it was shown that the higher the sulfur/oxygen ratio the larger the storage capacity as long as the sulfur amount did not exceed the optimum limit because otherwise conductivity might be negatively affected. It has been shown that that bio-based carbon electrodes in Li-Ss reduce the effect of the shuttle effect (Wang et al. 2017a; Senthil and Lee 2021). A recent study focused on designing high specific surface area and pore volume for a carbon anode, derived from reed flowers, in Li–S (Wang et al. 2020c). As a result of the tailored morphology, a large reversible specific capacity of 908 mAhg\(^{-1}\) with electrochemical stability over 100 cycles were achieved. It was argued that the large surface area and pore volume helped in increasing the adsorption of sulfur and polysulfide, thus minimizing dissolution of polysulfide or shuttle effect, and as a result large capacity good electrochemical stability were achieved. Other groups have also demonstrated the applicability of using the different properties of biomass-based electrodes to deliver high-performance in other storage batteries, e.g. as Li-OBs, NIBs, and KIBs, by manipulating the physical and chemical properties of those electrodes (Hao et al. 2018b; Sun et al. 2019b; Yang et al. 2020).

Efforts to develop high-performance composite biomass-based electrodes in storage batteries have also been reported (Zhang et al. 2020a; Sun et al. 2019b). Incorporating the right amount of NiCo\(_2\)O\(_4\) nanoparticles/nanoneedles in carbonized pomelo peel, working as anode in LIB, increased specific surface area that in turn enhanced storage capacitance up to 474 mAhg\(^{-1}\) after 120 cycles (@0.5 Ag\(^{-1}\)) (Zhang et al. 2020a). And also allowed for a stable performance over 1100 cycles at a capacitance of 363 mAhg\(^{-1}\) at current density of 2 Ag\(^{-1}\). In a separate study concerning the cathode of Li-Os, it was shown that by embedding CoFeP nanodots in a honeycomb-like anode derived from egg white rich with nitrogen and oxygen species, large storage capacitance of 1000 mAhg\(^{-1}\) after 141 cycles (@0.1 Ag\(^{-1}\)) was recorded as shown in Fig. 11 (Sun et al. 2019b). Although the incorporation of CoFeP nanodots might have blocked some pores on the surface of the carbonized egg white leading to a lower specific surface area than pristine carbonized egg white, the large capacitance and the
| Carbon precursor | Nanostructure of the electrode/Assembly state-of-the-art | Battery type | Electrolyte | Additive | Reversible specific capacity (mAh g⁻¹/# of cycles/ @current density (Ag⁻¹)) | Capacity retention rate (%) | Coulombic efficiency | References |
|------------------|---------------------------------------------------------|--------------|-------------|----------|------------------------------------------------------------------------------------------------|-----------------------------|-----------------------|-----------|
| Bagasse          | Honeycomb-like/Polymer binder                           | Lithium-ion  | 1.25 M LiPF₆ in a mixture of DMC<sup>a</sup> and EC<sup>b</sup> (1:1 vol%) | – | 691/100/0.1 – 99.1% after 50 cycles | Wan and Hu (2020) |
| Lignin           | Nanofiber membrane/Binder-free                          | Lithium-ion  | 1 M LiPF₆ in a mixture of EC &DEC<sup>c</sup> | Fe<sub>2</sub>O₃ | ~ 715/80/0.05 95.1% | Ma et al. (2019) |
| Lignin           | Nanofiber membrane/Binder-free                          | Lithium-ion  | 1 M LiPF₆ in a mixture of EC &DEC with 3 wt.% VC<sup>d</sup> | – | 611/500/[0.5C] – – | Culebras et al. (2019) |
| Pomelo peels     | Wrinkled sheet/Polymer binder                           | Lithium-ion  | 1 M LiPF₆ in a mixture of EC and DMC (1:1 vol%) | NiCo₂O₄ | ~ 500/120/0.05 – 97.6% after 120 cycles | Zhang et al. (2020a) |
| Corn straw       | Nanoparticles/Polymer binder                            | Lithium-ion  | 1 M LiPF₆ in a mixture of EC and DMC (1:1 vol%) | – | 546/100/[0.2C] – 85.8% after 100 cycles | Yu et al. (2020) |
| Seaweed          | 3D connected network/Binder-free                        | Lithium-ion  | 1 M LiPF₆ in a mixture of EC and DMC (1:1 vol%) | – | 550/300/[1C] – – | Zhang et al. (2017a) |
| Reed flowers     | Nanoporous powder/Polymer binder                        | Lithium-sulfur | 1 M LiTFSI in a mixture of DME<sup>a</sup> &DOL<sup>e</sup> (1:1 vol%) | – | 908/100/[0.1C] – 99.7% after 100 cycles | Wang et al. (2020c) |
| Pomelo peel      | Nanoporous powder/Polymer binder                        | Lithium-sulfur | 1 M Li2S &sublimated sulfur in a mixture of DME &DOL (1:1 vol%) | – | 718/300/[0.2C] – 98% after 300 cycles | Xiao et al. (2020) |
| Nanocellulose    | 3D nanofibril network/Binder-free                       | Lithium-sulfur | 1 M LiTFSI in a mixture of DME &DOL (1:1 vol%) | – | 590/200/[0.5C] – Close to 100% after 200 cycles | Chen et al. (2020a) |
| Egg white        | 3D honeycomb-like/Polymer binder                        | Lithium-oxygen | 1 M LiN(CF<sub>3</sub>SO<sub>2</sub>)<sub>2</sub> in a mixture of TEGDME<sup>d</sup> | CoFeP | 1000/141/0.1 – – | Sun et al. (2019b) |
| Carbon precursor        | Nanostructure of the electrode/Assembly state-of-the-art | Battery type                  | Electrolyte                        | Additive | Reversible specific capacity (mAhg$^{-1}$)/# of cycles/ @current density (Ag$^{-1}$) | Capacity retention rate (%) | Coulombic efficiency | References               |
|------------------------|----------------------------------------------------------|--------------------------------|-----------------------------------|----------|--------------------------------------------------------------------------------|----------------------------|-----------------------|-----------------------|
| Starch                 | Nanoporous skeleton/Polymers                             | Lithium-oxygen                | 1 M LiTFSI in a mixture of TEGDME | IrCo     | 100/ ~ 210/ 0.2                                                                 | 77                         | –                     | Shen et al. (2019)    |
| Pomelo peel            | Microtube/Coated on carbon paper                         | Lithium-oxygen                | 1 M LiCF3SO3 in a mixture of TEGDME | NiFe     | 13.8/290/0.1*                                                                | –                          | 99.4                  | Jing et al. (2019)    |
| Bagasse                | Honeycomb-like/Polymers                                  | Sodium-ion                    | 1.25 M NaPF6 in a mixture of DMC & EC (1:1 vol%) | –     | 506/100/0.1                                                                 | –                          | Close to 100% after 50 cycles | Wan and Hu (2020)    |
| Bacterial cellulose    | Nanofiber membrane/Binder-free                           | Sodium-ion                    | 1 M NaOTf in a mixture of diglyme | –       | 233/100/0.2                                                                  | –                          | –                     | Yang et al. (2019)    |
| Chitin                 | Nanofiber/Polymers                                        | Sodium-ion                    | 1 M NaClO4 in a mixture of EC & DEC (1:1 vol%) & 5 wt.% FEC | –     | 105/8000/1                                                                  | 85%                        | Close to 100% after 8000 cycles | Hao et al. (2018b)    |
| Spring onion peel      | Parallel thin flake/Polymers                              | Sodium-ion                    | 1 M NaClO4 in a mixture of EC & DEC (1:1 vol%) & 5 wt.% FEC | –     | 605/2000/0.05                                                                 | 94%                        | –                     | Zhao et al. (2020)    |
| Cotton linter pulp     | Wrinkled flake cauliflower/Polymers                       | Sodium-ion                    | 1 M NaClO4 in a mixture of EC and PC $^c$ (1:1 vol%) & 5 wt.% FEC | –     | 395/500/0.1                                                                 | –                          | Close to 100% after 500 cycles | Dan et al. (2020)     |
| Potato                 | Porous powder/Polymers                                    | Potassium-ion                 | 3 M KFSI$^b$ in a mixture of DME | –       | 248/100/0.1                                                                   | 91.7                       | Close to 100% after 400 cycles | Cao et al. (2019)     |
| Chitin                 | Nanofiber powder/Polymers                                 | Potassium-ion                 | 0.8 M KPF$_6$ in a mixture of EC & DEC (1:1 vol%) | –     | 215/100/ [0.2C]                                                             | –                          | 90% after 100 cycles | Hao et al. (2018a)    |
| Ganoderma lucidum spore (Fungus) | Cage-like porous powder/Polymers            | Potassium-ion                 | 1 M KPF$_6$ in a mixture of EC & DEC (1:1 vol%) | –     | 407/50/0.05                                                                  | 94.1                       | 90% after 50 cycles | Yang et al. (2020)    |
stability over 140 cycles were the result of unique physical structure (honeycomb-like) and the combined effect and activity role of Co, Fe, and P.

Most of the high-performance electrodes that are derived from some biomass precursors are assembled using polymer binders. Although polymer binders are essential additive to support the active powder in storage batteries, as a consequence, the dead-weight of the electrode increases, overall electrochemical performance drops, and the preparation time and cost increase (Zhang and Lou 2013; Shen et al. 2015; Zhang et al. 2017b). A common approach to tackle this issue is done by adopting self-standing materials such as in the case of lignin-based CNFs and bacterial cellulose-based CNFs. Table 7 summarizes the electrochemical performance of polymer binders and binder-free electrodes derived from a wide variety of biomass and biowaste materials. Using lignin as CNFs precursor to fabricate anode for LIBs, a remarkable reversible specific capacity of around 715 mAhg⁻¹ with capacity retention over 95% was reported, albeit the electrochemical stability extends to only 80 cycles (Ma et al. 2019). The large capacity was the result of the incorporation of iron oxide nanoparticles which have high theoretical capacity. Self-standing bacterial cellulose-based CNFs as an anode in NIB resulted in a somewhat moderate reversible specific capacity of 233 mAhg⁻¹ (Yang et al. 2019). However, most self-standing biomass-based carbon electrodes lack the ability of self-doping due to the nature of the starting materials, e.g. lignin, bacterial cellulose, and cellulose acetate, that luck enough nitrogen and oxygen species after carbonization. Hence, adopting an appropriate aerogel preparation protocol as the assembly method for the biomass materials that sustain self-doping is a promising approach and also promotes a unique physical property. For example, by adopting alginate from seaweed as the starting carbon precursor, a carbon aerogel anode was fabricated for Li–S (Zhang et al. 2017a). It was shown that even at high current density of 1C the reversible specific capacity reaches 550 mAhg⁻¹ after 300 cycles. The adoption of nitrogen- and oxygen-enriched species biomass- and biowaste-based carbon aerogel anodes in storage batteries is still limited and more research is needed to unravel its true potential.

High-performance bio-based nanostructured electrodes in supercapacitors

SC is another important class of storage devices that can deliver high power in short period of time. Their excellent cycling stability is another outstanding feature that storage batteries lack. The main parts of a supercapacitor are electrodes, electrolyte, and a separator. As in the case with rechargeable batteries, the electrolyte provides the medium for ions to float during charging and discharging, and the separator keeps the electrodes physically apart. The electrodes are the main functional part that contributes the most

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**Table 7 continued**

| Carbon precursor | Nanostructure of the electrode/Assembly state-of-the-art | Battery type | Electrolyte | Additive | Reversible specific capacity (mAhg⁻¹)/# of cycles/@current density (Ag⁻¹) | Capacity retention rate (%) | Coulombic efficiency | References |
|------------------|--------------------------------------------------------|--------------|-------------|----------|---------------------------------------------------------------------------------|-----------------------------|----------------------|------------|
| Bamboo charcoal  | Rod-like powder/Polymerr binder                        | Potassium-ion| 0.8 M KPF₆ in a mixture of EC & DEC (1:1 vol%) | –        | 393/300/0.05                                                                   | 87.1                        | Close to 100% after 300 cycles | Tian et al. (2020) |

*Capacity and current density units are in mAhcm⁻² and mAcms⁻², respectively. a refers to dimethyl carbonate, b refers to ethylene carbonate, c refers to diethyl carbonate, d refers to 1,2-dimethoxy ethane, e refers to 1,3-dioxolane, f refers to tetraethylene glycol dimethyl ether, g refers to sodium triflate, h refers to fluoroethylene carbonate, i refers to propylene carbonate, and j refers to potassium bis(fluoro-slufonyl)imid
to the electrochemical performance. One of the best suited electrodes is carbon-based due to their excellent physical properties and chemical stability in SCs (Merlet et al. 2012).

The recent research in SCs has been concerned about obtaining high-performance electrodes derived from renewable sources. Although the starting biomass and biowaste materials are ubiquitous, the approach of obtaining high-performance electrodes is almost the same which is fabricating electrodes with superior specific surface area, controlled physical structure, doping with different atoms, and enhanced

Fig. 10 Electrochemical and cycle performance of bagasse-derived electrodes at different current densities in (1) LIB and (2) in Li–S. Reprinted with permission from Wan and Hu (2020). Copyright (2020) Elsevier

Fig. 11 Electrochemical and cycle performance of CoFeP nanodots incorporated in a honeycomb-like electrode derived from egg white at different current densities. Reprinted with permission from Sun et al. (2019b). Copyright (2018) Elsevier
 pseudocapacitance activity by incorporating active materials. Electrodes made of activated eggshells, in a supercapacitor that its other parts (electrolyte and separator) are also made of the rest of the eggs (white, yolk, and membrane), delivered high capacitance of 421 F g\(^{-1}\) with high power of 10 KWkg\(^{-1}\) (Zhang et al. 2019b). The results were attributed to the high specific surface area and self-doped nitrogen, oxygen, and sulfur species that might have triggered pseudocapacitance activities. Utilizing walnut shell-based electrodes with ultrahigh specific surface area and tuned pore size delivered capacity of 216 F g\(^{-1}\) with high energy density and ultrahigh power density of 48 Whkg\(^{-1}\) and 100 KWkg\(^{-1}\), respectively (Shang et al. 2020). This is illustrated by Fig. 12 which shows a comparison between the electrochemical performance of the walnut shell-based electrode and an activated carbon counterpart. The large surface area facilitated ion and electron transport, while the tuned pores facilitated accumulation of charges, thus larger capacitance was achieved. Other recent efforts have utilized biomass and biowaste materials such as cabbage, durian, and aloe to obtain high-performance electrodes to deliver high power density with relatively high energy density (Cai et al. 2018; Yin et al. 2019; Lee et al. 2020). Table 8 summarizes the electrochemical performance of various bio-based carbon materials in SCs. The excellent electrochemical performance can also be explained by the inner structure that provides new paths for ion diffusions and space for storing charges, and enhanced interface between electrodes and electrolyte which is the result of nitrogen and oxygen species. In addition, the nitrogen content also increases electronic conductivity that in turn increases pseudocapacitance activities.

Unlike electrodes in batteries, binder-free electrodes in SCs are much common. The two most common approaches for a binder-free electrode are electrospun membranes and aerogels as summarized in Table 8. With such processed materials it is not only possible to obtain high-performance and self-standing materials, but also it is possible to modify the starting materials to embed additives to further enhance the performance. Incorporating graphene nanosheets in
Table 8  Electrochemical performance of biomass- and biowaste-based carbon electrodes in supercapacitors

| Carbon precursor                     | Nanostructure of the electrode/Assembly state-of-the-art | Electrolyte | Additive | Specific capacitance \( (\text{F g}^{-1})/\text{current density (A g}^{-1}) \) | Energy density \( \text{(Wh kg}^{-1}) \) | Power density \( \text{(W kg}^{-1}) \) | Rate capability | References               |
|-------------------------------------|----------------------------------------------------------|-------------|----------|--------------------------------------------------------------------------------|--------------------------------|-----------------|---------------------|---------------------|
| Organosolv lignin                   | Nanofibers/Binder-free                                    | 6 M KOH     | Graphene nanosheet                          | 267/1                             | 9.3                           | 493               | 97% after 5000 cycles | Dai et al. (2019)    |
| N. Enzymatic hydrolysis lignin      | Nanofibers/Binder-free                                    | 6 M KOH     | –        | 345/1                                           | –                             | –                | 97% after 2000 cycles | Zhang et al. (2020b) |
| Lignin extracted from poplar sawdust| Multichannel nanofibers powder/Binder-free              | 6 M KOH     | SnO_2     | 406/0.5                                         | 11.5                          | 451               | 95% after 10,000 cycles | Cao et al. (2020)   |
| Cellulose nanofibrils               | 3D nanofibril network/Binder-free                        | 2 M KOH     | NiCo_2S_4 | 1569/0.5                                        | 53.7                          | 184               | 78% after 5000 cycles | Liu et al. (2020a)  |
| Chitin                              | hierarchical nanoporous structure/Binder-free            | 6 M KOH     | –        | 413/0.5                                         | 9.7                           | –                | 99.6% after 10,000 cycles | Wang et al. (2020b) |
| Chitin                              | 3D nanofibril network/Binder-free                        | 6 M KOH     | –        | 221/1                                           | –                             | –                | 92% after 8000 cycles | Ding et al. (2018)  |
| Eggshell                            | Graphene-like nanoporous powder/Binder-free              | 6 M KOH     | mixed with egg white & yolk                  | 421/0.5                          | ~ 7                           | ~ 10,000          | –                   | Zhang et al. (2019b) |
| Walnut shell                        | Stacked flake powder/Binder                               | 6 M KOH     | –        | 216/0.5                                         | 48                            | 100,000           | 81% after 10,000 cycles | Shang et al. (2020) |
| Aloe                                | 3D net-like network/Binder-free                          | 6 M KOH     | Co_3O_4   | 1345/1                                          | 68.2                          | 549               | 92.7% after 10,000 cycles | Yin et al. (2019)   |
| Cabbage                             | 3D nanoporous network/Binder-free                        | 6 M KOH     | –        | 291/0.5                                         | 97.1                          | 1456              | 96.8% after 10,000 cycles | Cai et al. (2018)   |
| Durian                              | 3D tunneled network/Binder-free                          | 0.5 M KOH   | –        | 519/1                                           | 41.5                          | 730               | –                   | Lee et al. (2020)   |

A lignin-PAN-based electrospun electrode was argued to increase specific surface area and to anchor nitrogen and sulfur species in the base material after carbonization that otherwise might have decomposed to gases (Dai et al. 2019). The composite delivered an enhanced energy density of around 9.3 Wh kg\(^{-1}\) compared to the pristine electrode’s 4.1 Wh kg\(^{-1}\). Impressively, the incorporation of Co_3O_4 nanocrystals in aloe-based aerogel electrode improved the pseudocapacitance activity by delivering a capacitance of
1345 Fg\(^{-1}\) (\(\@1\) Ag\(^{-1}\)) with excellent cycling stability over 10,000 cycles and energy density of 68.2 WhKg\(^{-1}\) (Yin et al. 2019). Other additives such as SnO\(_2\) and NiCo\(_2\)S\(_4\) have been investigated for their performance in SCs (Liu et al. 2020a; Cao et al. 2020). Even with the substantial efforts that have been made to improve the current state of SCs more research is needed to deliver higher energy density at high power density of carbon electrodes derived from biomass and biowaste resources.

### Summary and perspective

Environmentally sustainable bio-based carbon precursors are excellent alternatives to precursors of petroleum chemicals origin. Bio-based carbon precursors are inexpensive due to their large abundance in nature. Moreover, bio-based carbon electrodes have excellent physical properties that permit them to perform outstandingly in energy storage applications. In this review, emerging and recent bio-based carbon precursors as high-performance electrodes in energy storage applications are classified and categorized based on their nanostructured morphologies (0D, 1D, 2D, and 3D) and natural origin. Some of these carbon precursors are lignin, cellulose, chitin, protein (e.g. egg), and other unconventional precursors (e.g. plant and food wastes). Under the four morphological categories, each of these materials is summarized and critically evaluated in terms of methodology, preparation protocols, morphology, physical properties, and doping mechanisms. In addition, the electrochemical performance of the most recent bio-based electrodes is discussed and summarized for rechargeable batteries and supercapacitors. The entirety of the review also serves as a compact guideline to design high-performance bio-based carbon electrodes for energy storage applications.

Tremendous efforts have been noted on designing and fabricating novel nanostructured bio-based electrodes with excellent morphological and physical properties that are capable of delivering high electrochemical performance in batteries and supercapacitors. Some studies focused on engineering nanoporous electrodes with specific surface area of larger than 1000 m\(^2\)/g and even exceeding 3000 m\(^2\)/g limit. Depending on the initial precursor, such physical properties are the results of the novel preparation protocols combined with some activation agents. While other studies worked on developing novel methods demonstrating the feasibility of heteroatoms- and self-doping in bio-based carbon electrodes. This is achieved either by finding methods to maintain the already existed species (e.g. nitrogen and oxygen groups) in the bone structure of the biomass material after carbonization, or by artificially importing some species by incorporating different substances. Such outstanding properties have shown to promote large capacity and ionic conductivity, increase in pseudocapacitance activity, and extend electrochemical stability. As a result, high-performance bio-based carbon electrodes have also been developed.

Despite the noted huge efforts to utilize bio-based carbon precursors as high-performance electrodes, more research focusing on further improving the current status of rechargeable batteries and supercapacitors to meet the ever-increasing future demands is imperative. To improve the status of bio-based carbon electrodes in energy applications the following challenges need to be addressed in future studies: (i) For most biomass and biowaste materials, the carbon yield is very low compared with synthetic materials. Increasing carbon yield would decrease the cost of processing which will reflect back on the total cost of the energy device. This can be approached by deploying additives that minimize the loss of carbon species during carbonization. (ii) The relation between specific surface area, pore size, and doping should be further investigated so that electrodes with optimum physical properties can be designed to obtain the best electrochemical performance possible. (iii) Due to the processing nature of some biomass and biowaste materials, the final carbonized state requires the use of organic binders to assemble it together as functional electrode. However, such approach undermines the overall performance of the electrode. To tackle this issue, developing new and simple preparation protocols to fabricate free-standing electrodes from materials that sustain self-doping are urged.

To summarize, environmentally sustainable bio-based carbon electrodes possess unique physical and chemical properties for high-performance electrochemical applications. Further research and more efforts are needed to tackle the immediate barriers to fully exploit the potential of the electrodes in energy storage applications.
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