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## Sustainably transforming biomass into advanced carbon materials for solid-state supercapacitors: a review

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Biomass-derived carbon materials (BDCMs) are widely considered as promising and practical candidates for electrode materials of solid-state supercapacitors (SSCs), due to their low cost, good chemical and mechanical stabilities, excellent electrical conductivity, and high deployment feasibility. Numerous investigations have recently been conducted for sustainably transforming biomass into electrode materials with high electrochemical performance in SSCs, even guided by data-driven approaches. Therefore, this review addresses conventional and emerging synthesis routes for BDCM-based electrode materials and discusses recent advances in energy storage mechanisms and electrochemical performance enhancement of BDCMs for SSCs, improving electrode preparation and performance optimization of BDCMs in a practical and efficient manner. As two of the most powerful tools for novel material discovery and design, machine learning (ML) and 3D printing technologies are introduced to provide closed-loop guidelines for accurately and efficiently producing BDCMs with excellent electrochemical performance; main challenges for successfully applying ML and 3D printing methodologies are also addressed, providing critical guidelines for potential innovation and future development of BDCM-based SSCs. In this review, from life-cycle perspective, environmental benefits are assessed for BDCM-based SSCs, being highlighted as a promising and practical alternative to solidify energy security and achieve sustainable biomass management. The concluding remarks and future prospects are finally discussed to provide valuable insights for academic researchers and governmental policymakers. With concerted efforts, sustainably transforming biomass into high-performance electrode materials for SSCs is beneficial to achieving UN Sustainable Development Goals 7, 11–13.

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## 1. Introduction

### 1.1 Current advances in energy storage technologies

The excessive use of traditional fossil fuels has caused irreversible damage to our living environment and entire ecosystem. Compared to traditional fossil fuels, renewable energy (*i.e.*, solar thermal and wind energy) has drawn significant attention. For example, the global installed capacity of renewable energy has continuously increased, with a growth rate of 115% during the last decade (2013–2022).<sup>1</sup> However, the inherent

characteristics of renewable energy result in instability and intermittency in the power generation process, making it difficult to match well with the demands of power grids. To overcome these existing obstacles, advanced energy storage technology has been widely considered as one of the most promising and practical approaches, strongly ensuring the stable operation of power grids and the reliability of power supplies.<sup>2</sup>

Current energy storage technologies mainly include pumped storage,<sup>3</sup> compressed air energy storage,<sup>4</sup> flywheel energy storage,<sup>5</sup> batteries,<sup>6</sup> fuel cells,<sup>7</sup> and supercapacitors (SCs).<sup>8,9</sup> As summarized in Table 1, both the advantages and disadvantages of these energy storage technologies are compared in detail. Numerous investigations have been performed on SCs, mainly due to their high-power density, rapid charge and discharge capability, and long cycle life. And incentive policies for accelerating the commercialization of SCs are globally developed as shown in Fig. 1. In 1996, the development of SCs was added as one core project of the ‘New Sunshine

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Table 1 Performance comparisons of different energy storage devices

|                         | Pumped storage | Compressed air | Flywheel | Battery (LEs) <sup>a</sup> | Fuel cell (LEs) <sup>a</sup> | SCs (LEs) <sup>a</sup> | SSCs       |
|-------------------------|----------------|----------------|----------|----------------------------|------------------------------|------------------------|------------|
| Energy density          | High           | Medium         | High     | Medium                     | High                         | Medium                 | Medium     |
| Power density           | Medium         | Medium         | High     | Medium                     | Medium                       | High                   | High       |
| Charge-discharge rate   | Slow           | Medium         | Fast     | Fast                       | Fast                         | Super-fast             | Super-fast |
| Production cost         | Medium         | Medium         | High     | High                       | High                         | Medium                 | Medium     |
| Reliability             | High           | Medium         | High     | Medium                     | High                         | High                   | High       |
| Cyclic stability        | High           | High           | High     | Medium                     | High                         | High                   | High       |
| Location constraints    | ✓              | ✓              | ✗        | ✗                          | ✗                            | ✗                      | ✗          |
| Construction period     | Long           | Medium         | Medium   | Short                      | Short                        | Short                  | Short      |
| Environmental benefits  | High           | High           | High     | Medium                     | Medium                       | High                   | High       |
| Application flexibility | Low            | Medium         | Medium   | High                       | High                         | High                   | High       |
| Pliability              | —              | —              | —        | Low                        | —                            | Low                    | High       |
| Electrolyte leakage     | —              | —              | —        | ✓                          | ✓                            | ✓                      | ✗          |
| Ref.                    | 10             | 11             | 12       | 13 and 14                  | 7, 15 and 16                 | 17 and 18              | 19–21      |

<sup>a</sup> LES stands for liquid electrolytes.

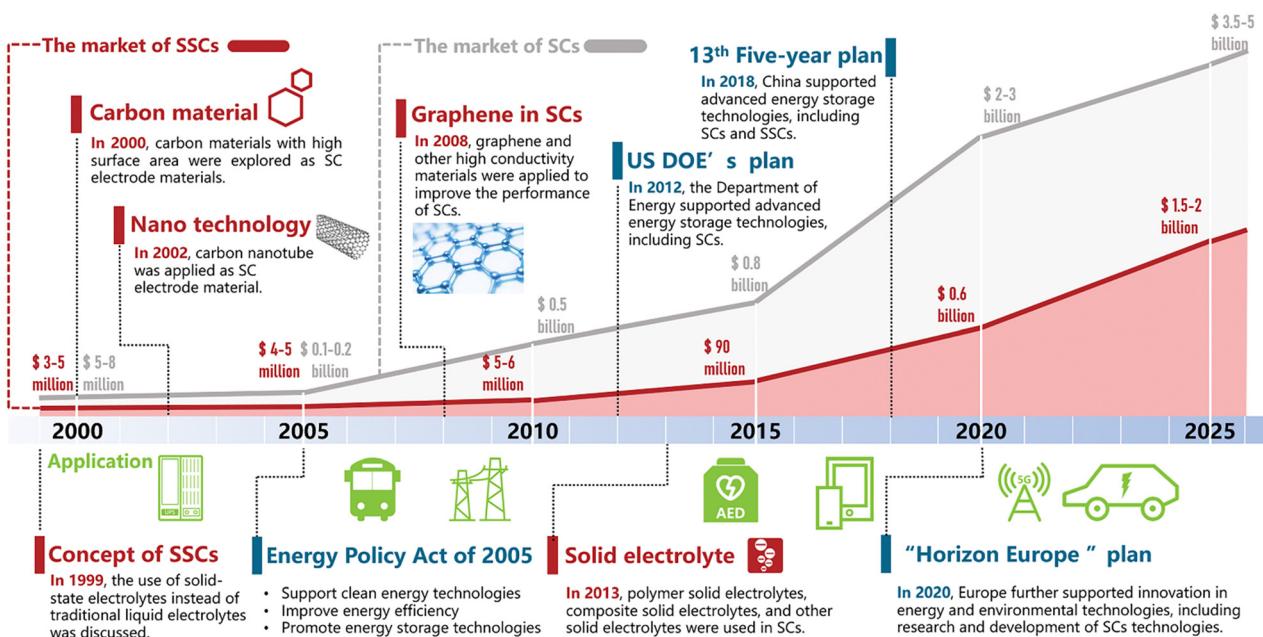


Fig. 1 A timeline of supercapacitor and solid-state supercapacitor development for solidifying energy security.

Program”, which was implemented in Japan. Scientists initiated the EU-funded project ‘new generation, high energy and power density SC based energy storage systems’ (HESCAP) to increase SCs’ energy density and make them cost-competitive with conventional batteries. In the US, the DOE’s (Department of Energy) Office of Electricity has announced funding opportunities, such as the \$27 million allocation to push emerging technologies like SCs from the lab into practical, scalable applications. In 2023, the “Guiding Opinions on Promoting the Development of Energy Electronics Industry” was promulgated to strengthen the industrialization of new energy storage devices (*i.e.*, activated carbon-supported SCs) in China.

## 1.2 Energy storage mechanism and electrolyte materials of SSCs

According to different charge storage mechanisms, SCs are divided into two main categories including electric double-layer capacitors (EDLCs) and pseudocapacitors. And they are

widely used in various fields, such as electronic devices,<sup>22</sup> transportation,<sup>23</sup> renewable energy storage,<sup>24–26</sup> industrial equipment,<sup>27</sup> medical devices,<sup>28,29</sup> and aerospace.<sup>30</sup> However, mainly due to their low energy density, SCs are still far from commercial applications. Liquid electrolyte-based SCs have poor flexibility and high electrolyte leakage risk, suggesting that more attention needs to be paid to their safety in use. This limits their applications in flexible electronics, flexible energy storage devices, and wearable technology.<sup>19,20,31,32</sup> Solid-state supercapacitors (SSCs) use solid materials as electrolytes. The energy storage mechanism of SSCs is similar to conventional SCs, both of them store charge by forming an electric double-layer or running rapid redox reactions, and it is worth noting that the main difference between SSCs and conventional SCs is the electrolytes.<sup>19,33</sup> In SSCs, ions in solid electrolytes are mainly conducted by point defects in the lattice, and their ionic conductivity is mainly affected by the material structure,

diffusion path and external conditions. However, in SCs, ions move freely in liquid electrolytes, and their conduction rate is mainly affected by ion concentration, temperature, and electric field intensity.

Solid-state electrolytes fundamentally eliminate the leakage risk associated with liquid electrolytes and display long life-spans and great adaptability to temperature variations, owing to that solid electrolytes typically exhibit excellent chemical and thermal stabilities. Electrolytes for SSCs mainly include the following four types: (1) polymer electrolytes in solid and gel forms; solid polymer electrolytes (SPEs) are mainly composed of polyethylene oxide (PEO) and salts (*i.e.*, LiCl) and gel polymer electrolytes (GPEs) are mainly composed of a polymer matrix (*i.e.*, polyvinyl alcohol), (2) inorganic solid electrolytes, commonly exhibiting poor flexibility but have high mechanical strength and thermal stability (*i.e.*, Li<sub>2</sub>S-PS<sub>5</sub>), (3) ionic liquid-based solid electrolytes, which are composed of ionic liquids and polymer matrixes, and (4) environmentally friendly gel polymer electrolytes, where renewable or biodegradable materials serve as the polymer matrix of the electrolyte (*i.e.*, corn starch and chitosan-supported electrolytes).<sup>34</sup> Therefore, SSCs are well-suited for application in flexible electronic devices, flexible energy storage devices, and wearable technology<sup>19,22,35</sup> and with the expansion of application scope, the market share is also growing, as shown in Fig. 1.

### 1.3 Biomass-derived carbon materials (BDCMs) for SSCs

Numerous investigations on the design and synthesis of novel solid electrode materials (*i.e.*, carbon-based materials, metal oxides, conductive polymers, and their composites) for SSCs have been performed for enhancing the energy density of SSCs. Among all solid candidates, carbon materials are considered as promising candidates for SSCs,<sup>36</sup> due to their cost-effectiveness, high stability, well-developed pore structure, and excellent electrical conductivity. As a representative of solid carbon materials, BDCMs have attracted widespread attention in the field of energy storage,<sup>37,38</sup> particularly in SSCs, mainly because BDCMs not only exhibit high stability and excellent conductivity but also present natural advantages of sustainable, abundant, cost-effective, and carbon-neutral properties. Here we would like to address the potential advantages of BDCMs used in SSCs as follows:

Firstly, as a sustainable and low-cost carbon precursor, biomass is valorised into advanced carbon materials using

one-pot or two-step approaches,<sup>39,40</sup> which is beneficial to achieving sustainable biomass management and a circular carbon economy, simultaneously. Compared to other carbon precursors, transforming biomass into carbon materials has advantages of higher cost-effectiveness and better environmental benefits. In addition, the used BDCMs are still environmentally friendly and available for soil remediations,<sup>41</sup> suggesting that the environmental impacts caused by waste BDCMs are relatively minimal, or even beneficial.

Secondly, after performing carbonization and activation, or even heteroatom-doping treatment, BDCMs display excellent textual properties and abundant active sites when compared with commercial activated carbons (shown in Table 2). Textural properties including the specific surface area (SSA) and pore volume are tunable and functional groups (*i.e.*, oxygen, nitrogen, and sulfur) are modifiable for achieving high-performance BDCM-based applications. Moreover, different biomass precursors significantly affect surface functional groups of BDCMs, and heteroatom-doping treatment is widely used for enriching more effective functional groups on the surface of BDCMs to further improve electrochemical performance.<sup>42</sup>

As summarised in Table 2, transforming biomass into carbon materials for electrode materials in SSCs is highly feasible, sustainable, and promising when compared with commercial carbon materials, owing to that BDCMs have key advantages of cost-effectiveness, environmental-friendliness, excellent pore structure, and favourable surface chemical properties. However, it is worth noting that different types of biomasses (*i.e.*, wood, crop residues, animal manure, *etc.*) possess distinct chemical compositions and physical structures, which are ultimately reflected in the resulting BDCMs. It suggests that selecting biomass precursors is the first and critical step for producing commercial-level BDCMs in SSCs. Moreover, the operating parameters, including operating temperature, heating rate, and activating agent, directly determine the textual properties of BDCMs. These could be well-solved by data-driven approaches including machine learning (ML), which is addressed in Section 3.

### 1.4 Challenges and opportunities

The previous reviews summarized in Table 3 have focused on material innovation, structural design, and performance

Table 2 The advantages and disadvantages of commercial carbon materials and BDCMs in SSC applications

|                             | Commercial carbon materials            | BDCMs   |
|-----------------------------|--|---|
| Production process          | Mature, large-scale production         | Immature, requires R&D and optimisation                   |
| Cost                        | Relatively high                        | Low   |
| Performance stability       | Stable                                 | Stable  |
| Specific capacitance        | Relatively low                         | High potential, can be enhanced through modification      |
| Environmental friendliness  | Moderate                               | Eco-friendly  |
| Electrical conductivity     | High                                   | High  |
| Chemical stability          | Good                                   | Good  |
| Operating temperature range | Wide                                   | Varies based on specific materials                        |
| Feedstock                   | Mineral resources, potentially limited | Biomass resources, abundant and diverse                   |
| Porosity                    | Moderate                               | High  |
| Doping treatment            | Limited                                | High potential  |
| Applications                | Widely used                            | High potential for expanded applications with further R&D |

Table 3 Literature reviews on novel materials for solid-state supercapacitors published from January 2014 to July 2024

| Electrode materials   | Performance indicators |                 |                 |                 |                 |                 |                  | 3D printing | Machine learning | Life-cycle assessment | Publication year | Total citations <sup>h</sup> | Ref.      |
|---|------------------------|-----------------|-----------------|-----------------|-----------------|-----------------|------------------|-------------|------------------|-----------------------|------------------|------------------------------|-----------|
|   | SC <sup>a</sup>        | CS <sup>b</sup> | CR <sup>c</sup> | ED <sup>d</sup> | PD <sup>e</sup> | BD <sup>f</sup> | LTP <sup>g</sup> |             |                  |                       |                  |                              |           |
| Conductive polymers, carbon-based materials, metal oxides, metal sulfides, and MXenes                   | ✓                      | ✓               | ✓               | ✓               | ✓               | ✓               | ✓                |             |                  |                       | 2014             | 744                          | 45        |
| Carbon-based materials, conductive polymers, composite materials, metal nitrides, and metal oxides      | ✓                      | ✓               | ✓               | ✓               | ✓               | ✓               | ✓                |             |                  |                       | 2014             | 1200                         | 46        |
| Poly(pyrrole) (PPy)   | ✓                      | ✓               | ✓               | ✓               | ✓               | ✓               | ✓                |             |                  |                       | 2016             | 662                          | 47        |
| Nanocarbon-based materials  | ✓                      | ✓               | ✓               |                 |                 |                 | ✓                |             |                  |                       | 2018             | 339                          | 35        |
| 2D material, metal oxides, metal nitrides, MOFs, conductive polymers, and carbon-based materials        | ✓                      | ✓               | ✓               | ✓               | ✓               | ✓               | ✓                |             |                  |                       | 2018             | 1284                         | 22        |
| Carbon-based materials, conductive polymers, metal oxides, metal sulfides, and MXenes                   | ✓                      | ✓               | ✓               | ✓               | ✓               | ✓               | ✓                |             |                  |                       | 2018             | 234                          | 48        |
| Polymer electrolytes  | ✓                      | ✓               | ✓               |                 |                 |                 |                  |             |                  |                       | 2019             | 2                            | 44        |
| Carbon-based materials, conductive polymers, and transition metal compounds (TMCs)                      | ✓                      | ✓               | ✓               |                 |                 |                 | ✓                |             |                  |                       | 2020             | 71                           | 31        |
| Graphene-based materials  | ✓                      | ✓               | ✓               | ✓               | ✓               | ✓               | ✓                |             |                  |                       | 2021             | 35                           | 32        |
| Carbon-based materials, conductive polymers, transition metal compounds (TMCs), and composite materials | ✓                      | ✓               | ✓               | ✓               | ✓               | ✓               | ✓                |             |                  |                       | 2022             | 50                           | 19        |
| COFs, MOFs, metal nitrides, MXenes, polyoxometalates (POMs), and black phosphorus                       | ✓                      | ✓               | ✓               | ✓               | ✓               | ✓               | ✓                |             |                  |                       | 2022             | 18                           | 20        |
| Graphene-based gels   | ✓                      | ✓               | ✓               |                 |                 | ✓               | ✓                | ✓           |                  |                       | 2023             | 33                           | 43        |
| Carbon-based materials, conductive polymers, MOFs, metal nitrides, metal oxides, and MXenes             | ✓                      | ✓               | ✓               |                 |                 | ✓               | ✓                |             |                  |                       | 2024             | 1                            | 49        |
| BDCMs   | ✓                      | ✓               | ✓               | ✓               | ✓               | ✓               | ✓                | ✓           | ✓                | ✓                     |                  |                              | This work |

<sup>a</sup> SC: specific capacity. <sup>b</sup> CS: cyclic stability. <sup>c</sup> CR: capacitance retention. <sup>d</sup> ED: energy density. <sup>e</sup> PD: power density. <sup>f</sup> BD: bending durability. <sup>g</sup> LTP: low-temperature performance. <sup>h</sup> Obtained from Web of Science (assessed by July 21, 2024).

optimization with cutting-edge technologies for accelerating commercial applications of SSCs. As displayed in Table 3, Lv *et al.*<sup>35</sup> reported the advantages of nanocarbon materials as flexible as SSC electrode materials, including their high SSA and excellent electrical and mechanical properties, summarized the latest application of nanocarbon materials in flexible SSCs systems and introduced many unique SSCs (*i.e.*, stretchable, self-healing, and shape-memory); Wu and Shao<sup>32</sup> introduced the ideal choice of graphene as an active electrode material for SCs, because it has extraordinary physical properties such as a large SSA, and reviewed two types of graphene-based all-solid-state supercapacitors (*i.e.*, film type and fibre type) in terms of the electrochemical/mechanical performances, strategies to improve the performance, and future perspectives. Zhang *et al.*<sup>43</sup> systematically introduced the preparation strategies and structural regulation methods of graphene-based gels and comprehensively addressed recent research progress in graphene-based gel composites for SSCs. The potential of graphene-based gels in enhancing the performance of flexible all-solid-state supercapacitors is emphasized, including their high-power density, excellent flexibility, and long cycle life. Additionally, the issue of a significant decrease in ionic conductivity at low temperatures, which leads to a reduction in energy density, is pointed out. The importance of improving the design, preparation, and performance enhancement of electrode and electrolyte materials is also proposed; Ainiya<sup>44</sup> summarized the latest research progress in solid-state electrolytes, including chitosan, polyacrylamide,

and polyethylene glycol, discussing the advantages and directions for performance improvement of these materials. Finally, despite some progress in the development of all-solid-state SCs, it should be noted that their performance still requires further optimization and development through performance enhancements in parameters (*i.e.*, ionic conductivity, energy density, and capacitance); Asl *et al.*<sup>19</sup> reviewed the advances in designing, fabricating, and characterizing flexible SSCs with high power density along with considerable rate performance, long cycle life, and high security, and several potential solid electrolyte materials and key parameters affecting the performance of SCs are introduced in detail; Dubal *et al.*<sup>22</sup> summarized the advancements in electrodes and electrolytes of SSCs and introduced some potential applications (*i.e.*, piezoelectric, photo-, shape-memory, self-healing). As summarized in Table 3, previous studies have primarily focused on material innovation, structural design, and performance optimization. However, the evaluation of SSCs has been limited to electrochemical performance, and especially the environmental benefits and economic feasibility are also missed in published reviews. As for ML, many previous studies have only focused on the concept of 'combining ML with material development' without systematically introducing ML technology for auxiliary material development. Additionally, there have been few reviews on the progress of BDCMs applied to SSCs, as shown in Table 3.

In recent years, the research directions of SSCs have a considerable shift from traditional material development and evaluation systems to efficient material development and

comprehensive evaluation systems with the goal of carbon neutralization. ML has already been increasingly applied on a laboratory scale to optimize both material design and process performance of BDCMs. The environmental impacts and feasibility of the production process of BDCM-based SSCs in industrial applications are still unclear, and it is necessary to comprehensively evaluate whether these processes meet the goal of carbon neutrality before deploying large-scale applications. Therefore, it is necessary to present a timely and comprehensive review of SSCs, focusing on recent advancements in the application of BDCMs in SSC electrodes and demonstrating the advanced technologies such as ML and 3D printing used to enhance the development and manufacturing of BDCMs. Additionally, this review provides a systematic environmental impact assessment from the life-cycle perspective and proposes the future developing directions of BDCMs and SSCs; aims to offer strong support for future research and application of SSCs, facilitating their advancement; and offers new perspectives and directions for future materials science research and the development of energy storage technologies.

## 2. Biomass-derived solid electrode materials for SSCs

Traditional SSC electrode materials are classified into two categories based on their energy storage mechanisms: (a) materials used for EDLCs are primarily carbon-based materials and their derivatives (*i.e.*, activated carbon, biochar, carbon nanotubes, and graphene),<sup>50–53</sup> and (b) materials used for pseudocapacitors include metal oxides/nitrides/sulfides and conducting polymers (*i.e.*, V<sub>2</sub>O<sub>5</sub>, RuO<sub>2</sub>, Fe<sub>2</sub>O<sub>3</sub>, TiN, VN, CuS, and PPy).<sup>47,54–60</sup> Metal oxide,<sup>61</sup> metal nitride,<sup>62</sup> and metal sulfide<sup>63</sup> electrodes in SCs typically exhibit faradaic pseudocapacitive effects, significantly enhancing capacitance. However, the high cost and relatively low stability have limited their widespread applications. Conducting polymers, due to their good electrical conductivity, have become an important choice for SC electrode materials. However, organic compounds are susceptible to environmental conditions, potentially causing permanent damage by volume expansion.<sup>64</sup> In addition, all the above-mentioned materials potentially cause environmental pollutions during material preparation and waste electrode disposal. As shown in Fig. 2, compared to the other two types of electrode materials, BDCMs have demonstrated strong vitality and competitiveness in the field of SSC electrode materials and becoming a hot topic of current research. Fig. 3 shows a common synthesis approach for transforming biomass into electrode materials for SSCs. Biomass precursors undergo initial pre-treatment including grinding and drying, followed by carbonization, to produce biochar materials. The biochar materials always have less developed pore structures, therefore, further activation and heteroatom-doping treatment need to be investigated for producing electrode materials with well-developed pore structures and effective functional groups, playing a critical role in achieving high-performance SSCs.<sup>65</sup>

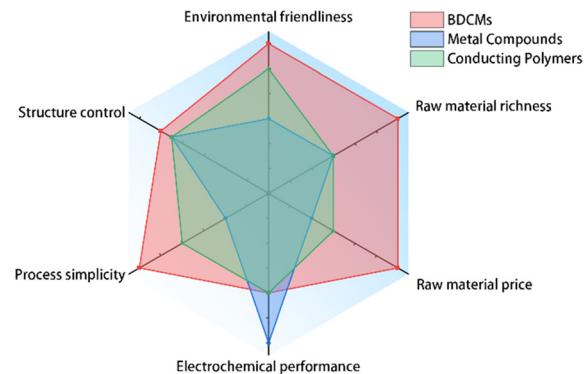


Fig. 2 Performance comparisons among different carbon precursor-derived electrode materials for solid-state supercapacitors.



Fig. 3 A common schematic diagram of developing BDCM-based SSCs.

The following part of this section will provide a detailed overview of the current research progress of BDCMs in SSCs.

### 2.1 Carbonization

Carbonization is a crucial step for producing BDCMs, significantly influencing their textural properties and thus affecting the electrochemical performance of SSCs. As displayed in Fig. 4 (red section), biomass carbonization, which is commonly termed pyrolysis, is the first step for valorizing biomass into carbon materials in the temperature range of 400–900 °C. When increasing pyrolysis temperature up to 200–400 °C, dehydration and devolatilization occur, where the water content and the volatile organic components (VOCs) are released to produce new porous structures or enlarge the existing pore structures.<sup>66</sup> With the increase of the heating temperature of 400–700 °C, biomass pyrolysis occurs where the macromolecules including cellulose, hemicellulose, and lignin are cracked into solid carbons (widely termed as biochar), gaseous products (*i.e.*, H<sub>2</sub>, CO<sub>2</sub>, CO, and CH<sub>4</sub>), and liquid products (*i.e.*, tar and oil).<sup>67</sup> When further increasing heating temperature over 650 °C,<sup>68</sup> the disordered structure in the carbon material begins to partially transform into a graphite-like structure, which is called the graphitization process.<sup>69</sup>

Hydrothermal carbonization (HTC) is a commonly used method for transforming biomass with a high-moisture content into carbon materials (widely termed as hydrochar), avoiding the pre-drying treatment for direct biomass carbonization.

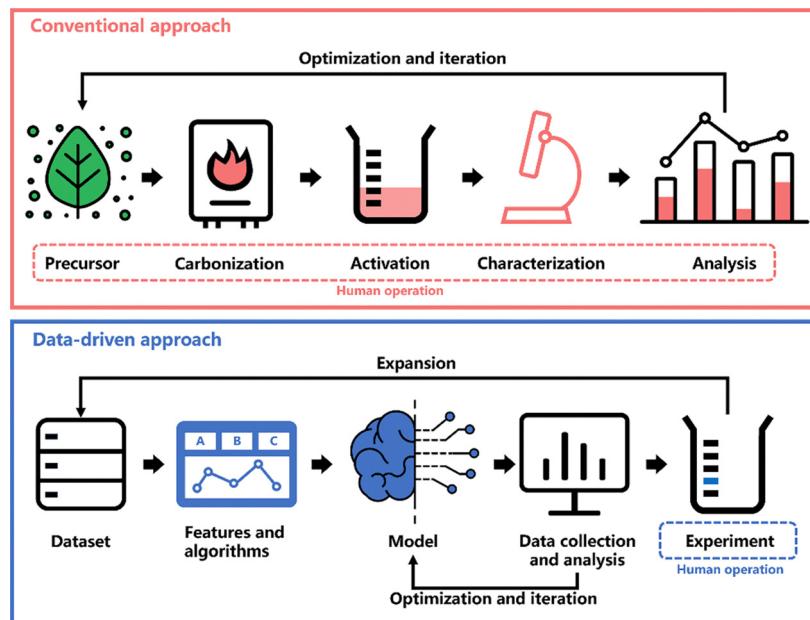


Fig. 4 Common schematic diagrams of conventional and data-driven syntheses of biomass-derived carbon materials.

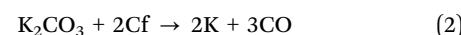
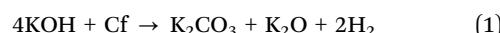
Xu *et al.*<sup>70</sup> conducted a simple HTC of crushed waste walnut shells at 180 °C, presenting a limited desired porous structure. Compared to HTC, direct carbonization requires higher temperatures with more energy consumption. Note that only HTC or direct carbonization is incapable of producing a well-developed porous structure.<sup>71</sup> It suggests that biochar and hydrochar are not listed as not good candidates for electrode materials, mainly due to their poor pore structures. Therefore, chemical and physical activation are used for further increasing the porosity of biochar/hydrochar, especially for microporous structures, which are beneficial to improving the electrochemical performance of SSCs when used as electrode materials.

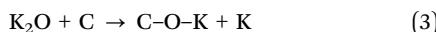
## 2.2 Activation

Activation aims to further enhance the SSA and optimize the pore structure of carbon materials by applying physical or chemical approaches, thereby improving their practical electrochemical performance (seen in Fig. 3).<sup>72</sup> Physical activation typically involves the partial gasification of the carbon framework in the presence of partially oxidizing media (*i.e.*, steam and CO<sub>2</sub>) or inert gases to develop new pores or slightly enlarge existing pores. At high temperatures, these activating agents react with the carbon framework to produce gaseous products (*i.e.*, CO and H<sub>2</sub>) to develop better pore structures.<sup>73</sup> Ahmad *et al.*<sup>21</sup> applied CO<sub>2</sub> activation for upcycling jujube into porous carbon materials at different activation temperatures. It suggests that the carbon material activated at 850 °C showed the best electrochemical performance, with a specific capacitance of 88.4 F g<sup>-1</sup> at 0.5 A g<sup>-1</sup>, and the assembled all-solid-state SCs exhibited an energy density of 9.6 W h kg<sup>-1</sup> and a power density of 87.86 W kg<sup>-1</sup>; when compared to commercial SSCs, they have competitiveness in high power density. Physical activation has the advantages of low cost, simplicity of operation, and

minimal pollution, however, its major drawback is the relatively low development of porosity after performing physical activation, resulting in relatively low pore volume and SSA. Additionally, high operating temperatures and long reaction times are required for effective physical activation.

Chemical activation is a widely used method for improving textural properties of BDCMs, using different chemical activating agents at high temperatures (500–800 °C).<sup>42</sup> The chemical activating agents are briefly categorized into alkaline activating agents (*i.e.*, KOH, NaOH, K<sub>2</sub>CO<sub>3</sub>, and Na<sub>2</sub>CO<sub>3</sub>) and acidic activating agents (*i.e.*, HNO<sub>3</sub>, H<sub>3</sub>PO<sub>4</sub>, and K<sub>2</sub>C<sub>2</sub>O<sub>4</sub>). Potassium hydroxide (KOH) and sodium hydroxide (NaOH) are two of the most effective chemical agents for developing microporous carbon materials.<sup>74</sup> Tobi and Dennis<sup>75</sup> prepared biomass-based activated carbons using the KOH agent. It was found that KOH-activated carbon materials exhibited a distinct honeycomb-like pore structure with a SSA of 915 m<sup>2</sup> g<sup>-1</sup>, which is bigger than that of physically activated carbons. Lee *et al.*<sup>76</sup> used renewable agricultural waste coconut shells as raw materials to prepare hierarchical activated carbon through HTC and KOH activation. The produced carbon materials exhibited a specific capacitance of 88 F g<sup>-1</sup> at 1 A g<sup>-1</sup> and an energy density of 48.9 W h kg<sup>-1</sup> at a power density of 1 kW kg<sup>-1</sup>, exhibiting a good capacitance retention rate of 92% even after 5000 cycles at 2 A g<sup>-1</sup>. The KOH activation mechanism of biomass-derived porous carbons mainly includes the following four steps.<sup>75</sup> During KOH activation, gasification reaction occurs in which KOH and K<sub>2</sub>CO<sub>3</sub> are reduced by carbon to K<sub>2</sub>O, K, CO, and CO<sub>2</sub>, which leads to the formation of a porous surface.





For NaOH activation, Norouzi *et al.*<sup>77</sup> prepared raw algal biochar (RAB) by direct pyrolysis of green macroalgae and further synthesized 3D interconnected mesoporous network algal biochar (3DFAB) through direct NaOH activation. The specific capacitances of symmetric SCs assembled were 158 F g<sup>-1</sup> for RAB and 296 F g<sup>-1</sup> for 3DFAB, respectively. The NaOH activation mechanism is elucidated that the formation of oxygen-containing functional groups is achieved through the rapid infiltration of OH<sup>-</sup> ions and NaOH into the pores of the biomass.

HNO<sub>3</sub> and H<sub>3</sub>PO<sub>4</sub> are widely used among all acidic activating agents, mainly due to that they increase porosity by dissolving cellulose<sup>78</sup> and also alter both numbers and types of surface functional groups on biochar.<sup>79</sup> Jiang<sup>80</sup> upcycled red cedar wood into carbon materials using HNO<sub>3</sub> activation, presenting that the specific capacitance of HNO<sub>3</sub>-activated biochar reached up to 115 F g<sup>-1</sup>. Yumak *et al.*<sup>81</sup> used switchgrass as the precursor and H<sub>3</sub>PO<sub>4</sub> as the chemical activator, and the SSA after activation increased to 1373 m<sup>2</sup> g<sup>-1</sup>, and the capacitive performance improved from 49 F g<sup>-1</sup> to 95 F g<sup>-1</sup>. These two activating agents increase both porosity and surface oxygen-containing functional groups to strongly enhance specific capacitance performance. In addition, zinc chloride (ZnCl<sub>2</sub>) is used as a chemical activating agent, and it is worth noting that heavy metal (Zn) pollution is unavoidable at the current stage. ZnCl<sub>2</sub> promoted the formation of charcoal and non-condensable gas by catalysing the breakage and recombination of chemical bonds during lignin pyrolysis and inhibited the formation of most phenolic monomers in bio-oil.<sup>82</sup> It suggests that ZnCl<sub>2</sub> enhanced the efficiency of lignin pyrolysis and the selectivity of products, resulting in the formation of carbon materials with a highly porous structure.

Compared to physical activation, chemical activation is more effective and efficient for producing carbon materials with a larger SSA and pore volume but has environmental drawbacks. After performing chemical activation, sample washing treatment needs to be conducted for removing extra chemical activating agents from the final prepared carbon materials, leading to secondary environmental pollution, especially for ZnCl<sub>2</sub>. Therefore, environmental impacts caused by chemical activation need to be well-considered in future research.

### 2.3 Heteroatom-doping treatment

Heteroatom-doping treatment is a common method for adding more effective functional groups on the surface of BDCMs, which are beneficial to further improving their electrochemical performance.<sup>83</sup> Due to the difference in bond energy between heteroatoms and carbon atoms, heteroatom-doping often introduces structural defects and active sites for improving the chemical reactivity of BDCMs. Heteroatom-doping treatment changes the surface chemical characteristics, strongly

enhancing the contact efficiency between the electrode and the electrolyte and promoting rapid ion transportation. In addition, some atoms (*i.e.*, N, B) provide additional electrons or generate holes (positive charge carriers), which can optimize the electronic conductive network of the material and improve the conductivity of BDCMs.<sup>83</sup>

Among all heteroatom-doping, N-doping is the most widely used for increasing effective functional groups of BDCMs. Chen *et al.*<sup>84</sup> used soybean milk as the sole carbon and nitrogen source, CaCO<sub>3</sub> nanospheres as the hard template, and KOH as the chemical activating agent to prepare N-doped hierarchical porous carbon nanospheres (NPCNs). As a result, NPCN exhibits a high SSA of 1208 m<sup>2</sup> g<sup>-1</sup>, a specific capacitance of 240.7 F g<sup>-1</sup>, and an excellent capacitance retention rate of 90% in 20 A g<sup>-1</sup>. The SSC assembled by NPCN showed an energy density of 10.2 W h kg<sup>-1</sup> in 351 W kg<sup>-1</sup>, which is superior to normal commercial SSCs. Traditional EDLCs have low energy density, and heteroatom-doping treatment clearly improved surface wettability and conductivity, enhancing the overall electrochemical performance.<sup>84</sup> In addition, dual-doping is developed for improving electronic conductivity and electrochemical activity of BDCM-based electrode materials based on synergistic effects.<sup>85</sup> Wu *et al.*<sup>86</sup> successfully prepared porous activated carbon (RFAC) derived from prickly pear waste residues by *in situ* nitrogen and oxygen-doping treatment. The optimized RFAC-6 exhibited an ultra-high SSA of 3952.9 m<sup>2</sup> g<sup>-1</sup> and was rich in heteroatom functional groups, demonstrating a remarkable specific capacitance of 370 F g<sup>-1</sup> at 0.5 A g<sup>-1</sup> and retained 95% of its capacitance after 5000 cycles. Geng *et al.*<sup>87</sup> utilized spirulina as a carbon and nitrogen source and employed KOH as an activating agent to synthesize a porous carbon material, which exhibited a high SSA of 2923.7 m<sup>2</sup> g<sup>-1</sup> and abundant heteroatoms, including O (13.78%) and N (2.55%). Notably, the material demonstrated an exceptional specific capacitance of 348 F g<sup>-1</sup>.

Apart from activation and heteroatom-doping treatment, researchers have innovatively developed other preparation techniques for BDCMs. Citric acid (CA) can cross-link with the cellulose on the surface of straw biomass channels to form a three-dimensional network structure. This enhances the load-bearing capacity of the straw biomass channels, facilitating the entry of activating reagents into the carbonized product after high-temperature carbonization. This process results in an excellent porous structure in the final carbon material. Du *et al.*<sup>53</sup> used wheat straw as the raw material and successfully prepared porous carbon materials through citric acid cross-linking and KOH activation. The optimal porous carbon derived from wheat straw was then assembled into SCs for testing electrochemical performance. The specific capacitance reached up to 294 F g<sup>-1</sup>, with a capacitance retention rate of 97.6% after 5000 cycles at 10 A g<sup>-1</sup>. The assembled SCs exhibited an energy density of 14 W h kg<sup>-1</sup> and a power density of 440 W kg<sup>-1</sup>. More importantly, the SCs were bent at 0°, 45°, 90°, and 135° and the CV curves almost fully overlapped, which revealed the outstanding flexibility. The assembled SC exhibited excellent EDLC performance even at temperatures as low as 20 °C. To further enhance

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electrochemical performance and overcome the inherent limitations of carbon materials, researchers have been mixing carbon materials with other materials to create composites. Hu *et al.*<sup>88</sup> prepared rice husk carbon (RHC) *via* calcination and grew a layer of nickel-cobalt double hydroxide (Ni Co-LDH) on the RHC surface using a hydrothermal method, achieving NiCo-LDH@RHC composites with a nanoflower structure. This approach not only introduced pseudocapacitive materials but also addressed the poor conductivity issues caused by the layered aggregation and stacking of nickel-cobalt double hydroxide. The nanoflower structure of the composite contains numerous active sites, allowing for more faradaic redox reactions.

In addition to high performance and long lifespan, the green and low-carbon synthesis processes of electrode materials should be highlighted as one of the important research directions. Wang *et al.*<sup>89</sup> proposed a simple, low-cost, and environmentally friendly method to synthesize high-performance hierarchical porous biochar (HPB) from gallnut (GC) for use in highly sustainable and recyclable all-solid-state in-plane micro-supercapacitors. This method avoids the use of any toxic and corrosive reagents, and the synthesized HPB demonstrated excellent cycling stability with a capacitance retention rate of up to 90% after 1000 cycles. Zhang *et al.*<sup>90</sup> constructed flexible SSCs using materials entirely derived from eggs. The eggshell and egg white/yolk were used to build the electrodes, the egg white/yolk was used to prepare the gel-like solid electrolyte, and the eggshell membrane was used as the separator. The activated egg-derived carbon (AEC-0.3) electrode exhibited a specific capacitance of  $420.8 \text{ F g}^{-1}$  at  $0.5 \text{ A g}^{-1}$  and retained 80% capacitance after 5000 cycles at a current density of  $1 \text{ A g}^{-1}$ . The assembled SCs demonstrated excellent flexibility, and the specific capacitance and the CV curve are almost unchanged when bending or twisting the SC.

BDCM-based SSCs have many advantages, however, their future commercialization is still restricted by the following shortcomings: (1) poor interface electrochemical performance between electrodes and solid electrolytes. Solid electrolytes are harder than liquid electrolytes. It is difficult for solid electrolytes to have full contact with electrodes, leading to a low effective contact area and low specific capacitance. From a long-term perspective, problems such as poor contact and increased interface resistance are prone to occur under this unstable contact condition. As for production, because the contact between electrodes and electrolytes needs to be ensured the assembly process will be more difficult than normal SCs, which may lead to high production costs; (2) low ionic conductivity. In solid electrolytes, the velocity of ionic conductivity is lower than that of liquid electrolytes, restricting the fast charge and discharge capacity and decreasing the power density. Especially under low-temperature conditions, the worse ionic conductivity will further limit the application of SSCs under extreme environments; and (3) limitation caused by biomass on large-scale BDCMs-based SSC applications. On the one hand, biomass feedstocks are significantly influenced by factors such as species origin, regionality, and even seasonality. Given that SC electrode materials are high-value

products, there is a compelling rationale to select high-quality raw materials (such as coconut shells and pine wood) to ensure product quality and uniformity. On the other hand, the treatment process of biomass materials (*i.e.*, pre-treatment, pyrolysis, activation, and heteroatom-doping) is highly energy-intensive and leads to environmental pollution. For example, the  $\text{ZnCl}_2$  chemical used during activation inevitably causes heavy metal pollution to our water system and finally our entire ecosystem. Therefore, achieving efficient recycling and reuse of these chemicals represents another challenge in the large-scale conversion of biomass into carbon materials.

### 3. Cutting-edge technical approaches for accelerating the commercialization of solid-state supercapacitors

#### 3.1 Machine learning

The conventional routes for new material discovery face multiple challenges. Firstly, material synthesis routes are commonly both labour- and time-consuming, resulting in a limited dataset for performance enhancement and material optimization. The high-throughput material synthesis techniques alleviate this issue to some extent, but their implementation costs remain very high. Secondly, the complexity and diversity of emerging material development make the traditional trial-and-error approach highly inefficient and difficult to optimize specific properties accurately. Therefore, data-driven approaches have been attracted significant attention for discovering emerging and advanced materials, owing to their advantages of improved accuracy, efficient optimization, and continuous improvement.

As one typical data-driven approach, ML algorithms extract valuable information from large datasets to design and optimize novel material discovery in an efficient manner.<sup>91</sup> To solve the inherent disadvantage of conventional trial-and-error approaches, ML-aided approaches are capable of revealing the underlying relationships among textural properties, functional groups, and application performance.<sup>92</sup> Moreover, high-performance BDCMs are accurately designed by subsequent ML model training and experimental revalidation.<sup>93,94</sup> ML, with powerful data processing capabilities, has brought revolutionary changes to the field of conventional material discovery for different research areas and academic disciplines, mainly including (a) ML-assisted material characterizations: data-driven methods automate characterization, accelerating data processing and reducing systematic errors. This enables high-throughput analysis with minimal human intervention. ML excels at handling complex, high-dimensional data, uncovering valuable insights, and reducing human workload;<sup>95</sup> (b) ML prediction of material properties: ML replaces costly first-principles simulations, enabling high-throughput material screening. By training ML models, multiple material performance indicators can be predicted, such as thermal and electrical conductivity. This approach aids in discovering new materials and optimizing existing ones;<sup>95</sup> (c) ML-assisted material synthesis: ML enhances material synthesis by integrating

intrinsic and extrinsic descriptors, and ML creates predictive models to guide the experiments effectively. This accelerates and optimizes the materials development<sup>95</sup> and (d) ML-assisted paradigm discovery: ML enhances scientific research by discovering new paradigms and validating existing ones through advanced data analysis. It identifies hidden patterns and relationships in large datasets, leading to faster hypothesis generation and testing.<sup>95</sup>

In addition, Fig. 4 represents schematic diagrams of conventional and data-driven syntheses of BDCMs, respectively. In the conventional approach, researchers need to extensively repeat the experiment–characterization–analysis cycle to optimize the materials. In the data-driven approach, a model is built and preliminarily analysed based on the existing dataset, and a small number of experiments need to be carried out for efficiently validating and optimising ML models. The collected data can expand the existing dataset to further optimize and iterate ML models, and finally achieve accurate performance optimization with less experiments and human operations.

The electrochemical performance is significantly affected by textural properties and functional groups of BDCMs, which are controlled by sample synthesis (*i.e.*, carbonization and activation methods). Therefore, how to quickly and accurately design synthesis routes of BDCMs with excellent electrochemical performance is crucial to accelerate commercial applications of SSCs. Mathew *et al.*<sup>96</sup> valorised jackfruit seeds into solid carbon samples for electrode materials, and also developed ML models for predicting specific capacitance and equivalent series resistance. The optimized jackfruit seed-derived carbon material, a well-developed porous carbon with a unique tubular morphology, exhibits high capacitance and low resistance in SSC applications. Sun *et al.*<sup>97</sup> listed 14 key parameters, including operating parameters of carbonization and activation and main indicators of electrochemical performance as the input and target features of ML algorithms. And they verified the gradient boosting regression (GBR) model with the best predictive performance and concluded that the heating rate is the most significant factor affecting the specific capacitance of BDCMs, with a negative correlation. Liu *et al.*<sup>98</sup> applied 6 ML models to predict the capacitance performance of BDCMs and concluded eXtreme Gradient Boosting (XGBoost) as the best prediction model. And the main results indicated that the surface area of micropores ( $S_{\text{micro}}$ ) and the ratio of  $S_{\text{micro}}$  to the SSA ( $S_{\text{micro}}/\text{SSA}$ ) had the most significant contributions to capacitance performance. Jha *et al.*<sup>99</sup> verified the artificial neural network (ANN) as the best model to predict the electrochemical performance of lignin-based SCs. Based on the above-mentioned results, at the current stage, researchers need to build multiple ML models to predict relationships between the synthesis parameters and the characteristics of BDCMs and between characteristics of BDCMs and electrochemical performances, separately.

As a powerful tool, ML changes the process of material development, but it still has limitations and challenges that should be addressed. First, the use of ML in materials science faces the problem of the lack of sufficient datasets for model

training and test.<sup>100</sup> Experimental data in materials science are usually very scarce, especially for new materials, the process of obtaining experimental and simulated data is often time-consuming and expensive, resulting in insufficient training data. Worse is that most of the data used in ML are collected manually. Human factors such as personal habits and personal preferences will cause human errors, and these collected data are very easy to manipulate at any stage before they are applied to ML algorithms. Therefore, a large amount of data often has quality-related issues, such as insufficient metadata descriptions, inconsistent document formats, date duplicates, date errors, which pose difficulties for researchers in data reuse and understanding. Second, many ML models (especially deep learning models) have ‘black box’ characteristics, and it is difficult to explain their internal working principles, which hinders researchers’ understanding of material behaviours and underlying physical and chemical mechanisms.<sup>101</sup> Therefore, it is difficult to extend this model to different material systems or different application scenarios. Third, in the field of carbon material development, the application of ML is currently focused on the prediction of material properties. It is more important to focus on the design of new materials, optimization of the manufacturing process, material characterization and analysis in future research.

Currently, data are mainly collected in a manual manner, which poses several pivotal challenges: (1) the small-scale dataset collected manually leads to a lack of generalization capability for ML algorithms, (2) manual data collection is prone to data missingness, errors, and biases, thereby affecting the accuracy and fairness of machine learning-aided investigations, and (3) manual data collection is time-consuming and labour-intensive, limiting the large-scale deployment of ML-guided practical applications.<sup>102</sup> In order to build the database accurately and swiftly, it is imperative to implement automated programs, including large language models<sup>103</sup> and web crawlers,<sup>104</sup> and for data collection it lies in their abilities to efficiently and accurately process and analyse vast amounts of data with high scalability and real-time capability. This significantly enhances both the efficiency and quality of data collection while reducing labour- and time-consumption. However, these approaches also pose challenges of data duplication and inaccuracies as well as the risk of violating copyright laws, when data are scraped directly from websites.<sup>104</sup> Hence, the development of legitimate and highly reliable automated data acquisition programs or software is imperative. Optimizing algorithms and rule designs, as well as incorporating manual review mechanisms, holds great significance in improving data accuracy. Prior to utilizing automated programs for data collection, thoroughly understanding relevant laws and regulations, as well as website agreements, effectively mitigates legal risks.

### 3.2 3D printing

3D printing, also known as additive manufacturing, constructs models through layer-by-layer material deposition, which is digitally controlled by computer-aided design (CAD) models.<sup>105</sup> Based on the material deposition method, curing technology, and

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the bonding mechanism, 3D printing technologies are categorized into 7 types: material extrusion, material jetting, vat photopolymerization, sheet lamination, powder bed fusion, directed energy deposition, and binder jetting.<sup>106,107</sup> 3D printing has been considered as one emerging route for printing high-performance electrode materials in the field of SCs.<sup>108</sup> At the current stage, 3D printing technologies are mainly used for printable materials (*i.e.*, electrodes, electrolytes, and current collectors), and post-processing, significantly enhancing the overall electrochemical performance of SCs.<sup>109</sup>

Preliminarily, 3D printing can be applied to the direct preparation of BDCMs. The biomass precursor is fabricated into the desired structure using 3D printing technology and then subjected to pyrolysis and activation steps to be converted into carbon materials. The carbon materials are finally assembled into SSCs. Shao *et al.*<sup>110</sup> used a blend of microfibrillated cellulose/lignosulfonate as the carbon precursor and 3D-printed rectangular gels. The gels were then subjected to high-temperature carbonization under an inert gas to obtain highly conductive carbon materials. 3D printing technology imposes stringent requirements on the properties of the materials being printed, and biomass precursors must undergo special treatment before they can be printed. Blyweert *et al.*<sup>111</sup> combined acrylic resin with a bio-based carbon precursor to create a composite resin, which was then used to construct material modules using stereolithography technology. The carbonized material exhibited excellent mechanical properties and thermal stability. This novel method for preparing carbon materials is highly suitable to produce electrode materials. Furthermore, components of SSCs such as electrodes and electrolytes can be manufactured using 3D printing technology and assembled in a single process. Idrees *et al.*<sup>112</sup> used porous carbon derived from packaging waste and gel electrolytes, employing 3D printing to shape the electrodes and electrolytes, thereby fabricating SSCs without any post-processing steps.

3D printing has many advantages in the field of advanced material production. First, 3D printing has design flexibility and can realize complex structure production through CAD, which is particularly useful for customized production and development of BDCMs with special shapes or functions. Second, 3D printing can quickly produce samples, shorten the material development cycle, and promote the development and commercialization of new BDCMs. At the same time, compared with the traditional manufacturing process, 3D printing technology uses the method of adding materials layer by layer, which can greatly reduce waste during the production of special-shaped materials. However, many challenges limit the application of 3D printing in the field of BDCM production. The biomass precursor materials suitable for 3D printing are limited, and the chemical compositions and physical properties affect the printing quality and the performance of the final product. Some 3D-printed BDCMs may not be as good as carbon materials prepared by traditional methods in mechanical properties, which may limit their use in some applications that require high strength or durability.

## 4. Sustainability of solid-state supercapacitors using biomass-derived carbon materials

In the context of carbon neutrality, the environmental benefits of emerging technologies and/or processes need to be fully assessed from a life-cycle perspective.<sup>65</sup> Therefore, life-cycle assessment (LCA) is applied to provide a comprehensive and systematic evaluation of BDCMs in SSCs for academic researchers and governmental policymakers. Commonly, the functional unit is defined as 1 kg electrode material. Three different system boundaries are shown in Fig. 5, and in this work, we choose the “cradle-to-gate” as a system boundary to assess the environmental benefits of five different electrode materials in SSCs, including (a) soybean-derived nitrogen-doped porous carbon (NPC),<sup>84</sup> (b) palm waste-derived porous carbon (PWPC),<sup>75</sup> (c) calcium-doped binary strontium-copper oxide (SCCO),<sup>113</sup> (d) Ni<sub>2</sub>P,<sup>114</sup> and (e) graphene oxide/polypyrrole (GO-PPy).<sup>115</sup> The lab-scale data on feedstock treatments, energy consumptions, and greenhouse gas emissions to produce these materials are collected from the corresponding references. In this study, the environmental impacts associated with each step were evaluated using the open-access LCA software OpenLCA 2.2.0 based on the Ecoinvent database v3.9 to obtain the background data. The impact assessment method used was CML v4.8 2016.

Fig. 6 compares the environmental impacts of five different solid materials-based SSCs, and more detailed comparisons are listed in Table 4. NPC exhibits significant advantages in terms of lower ecological impact and preparation cost, mainly due to the use of more environmentally friendly and sustainable biomass materials as well as the rare use of chemical reagents and low-energy consumption. For PWPC, although it uses palm waste as the raw material, its environmental impact is higher than NPC, mainly caused by the higher energy consumption of the preparation process. Therefore, the development of a promising and sustainable manufacturing process is one of the most important directions for the future development of BDCMs. The metal-based materials SCCO and Ni<sub>2</sub>P have relatively high carbon emissions and environmental impacts among these five solid materials. Compared with the low-cost and carbon-negative BDCMs, the processing of metal-based materials is more complex and requires higher energy consumption during the processing, and the price of the prepared electrode materials is also higher. The environmental impacts

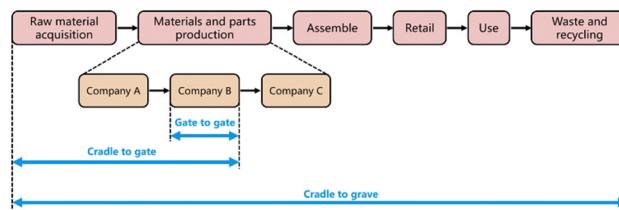


Fig. 5 A schematic diagram of common system boundaries used in LCA.

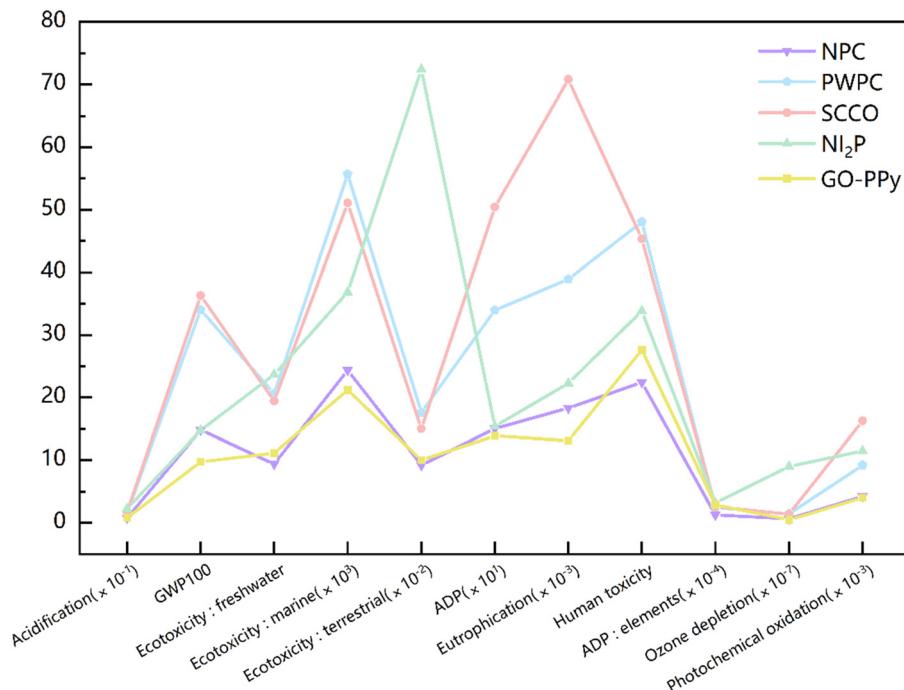


Fig. 6 Comparisons of environmental impacts of five different solid materials for SSCs.

Table 4 Detailed assessment of environmental impacts and economic feasibilities for five different solid material based SSCs

| Categories                                | NPC                   | PWPC                  | SCCO                  | Ni <sub>2</sub> P     | GO-PPy                | Unit                   |
|---|-----------------------|-----------------------|-----------------------|-----------------------|-----------------------|------------------------|
| Acidification                             | $7.82 \times 10^{-2}$ | $1.66 \times 10^{-1}$ | $1.73 \times 10^{-1}$ | $2.25 \times 10^{-1}$ | $8.02 \times 10^{-2}$ | kg SO <sub>2</sub> -Eq |
| Global warming potential (GWP100)         | $1.48 \times 10^1$    | $3.40 \times 10^1$    | $3.63 \times 10^1$    | $1.48 \times 10^1$    | 9.74                  | kg CO <sub>2</sub> -Eq |
| Freshwater aquatic ecotoxicity            | 9.39                  | $2.06 \times 10^1$    | $1.95 \times 10^1$    | $2.37 \times 10^1$    | $1.11 \times 10^1$    | kg 1,4-DCB-Eq          |
| Marine aquatic ecotoxicity                | $2.44 \times 10^4$    | $5.57 \times 10^4$    | $5.11 \times 10^4$    | $3.69 \times 10^4$    | $2.12 \times 10^4$    | kg 1,4-DCB-Eq          |
| Terrestrial ecotoxicity                   | $9.22 \times 10^{-2}$ | $1.76 \times 10^{-1}$ | $1.50 \times 10^{-1}$ | $7.24 \times 10^{-1}$ | $1.00 \times 10^{-1}$ | kg 1,4-DCB-Eq          |
| Abiotic depletion potential: fossil fuels | $1.50 \times 10^2$    | $3.39 \times 10^2$    | $5.04 \times 10^2$    | $1.54 \times 10^2$    | $1.39 \times 10^2$    | MJ                     |
| Eutrophication                            | $1.83 \times 10^{-2}$ | $3.89 \times 10^{-2}$ | $7.08 \times 10^{-2}$ | $2.23 \times 10^{-2}$ | $1.31 \times 10^{-2}$ | kg PO <sub>4</sub> -Eq |
| Human toxicity                            | $2.24 \times 10^1$    | $4.81 \times 10^1$    | $4.53 \times 10^1$    | $3.39 \times 10^1$    | $2.76 \times 10^1$    | kg 1,4-DCB-Eq          |
| Abiotic depletion potential: elements     | $1.27 \times 10^{-4}$ | $2.44 \times 10^{-4}$ | $2.56 \times 10^{-4}$ | $3.19 \times 10^{-4}$ | $2.90 \times 10^{-4}$ | kg Sb-Eq               |
| Ozone layer depletion                     | $6.55 \times 10^{-8}$ | $1.42 \times 10^{-7}$ | $1.39 \times 10^{-7}$ | $9.00 \times 10^{-7}$ | $4.61 \times 10^{-8}$ | kg CFC-11-Eq           |
| Photochemical oxidation                   | $4.21 \times 10^{-3}$ | $9.22 \times 10^{-3}$ | $1.63 \times 10^{-2}$ | $1.15 \times 10^{-2}$ | $4.00 \times 10^{-3}$ | kg ethylene-Eq         |
| Specific capacitance                      | 301                   | —                     | 308                   | 354                   | —                     | F g <sup>-1</sup>      |
| Cost                                      | 4+                    | 8+                    | 40+                   | 50+                   | 120+                  | USD \$                 |

NPC: soybean-derived nitrogen-doped porous carbon. PWPC: palm waste-derived porous carbon. SCCO: calcium-doped binary strontium-copper oxide. GO-PPy: graphene oxide/polypyrrole.

of the composite material GO-PPy are similar to those of NPC, but its production cost is much higher than that of NPC.

Traditional SC materials (*i.e.*, rare metal-based materials and polymers) are widely used in the electronics industries but are constrained by their high cost, high purity requirements, and resource limitation. Metal-based electrode materials, particularly rare metal-based, incur high production costs due to scarcity and high purity requirements, and polymer-based electrode materials need the addition of expensive additives and complex post-processing techniques to achieve excellent electrochemical performance. This suggests that more sustainable and low-cost electrode materials need to be synthesized as promising alternatives.

Transforming biomass into carbon materials for replacing traditional electrode materials in SSCs has its unique potential,

mainly owing to that biomass is a sustainable and low-cost carbon precursor. Moreover, as displayed in Fig. 6, BDCMs have demonstrated excellent environmental benefits in SC applications, with treated carbon materials even rivalling advanced metal materials in performance. SCs assembled from these cost-effective and high-performance materials are highly competitive in the market. From a perspective of economic feasibility, as listed in Table 4, the unit price of low-end SCs may be between a few dollars and a dozen dollars, while the price of high-end industrial or professional SCs can reach dozens of dollars or even higher. Taslim *et al.*<sup>116</sup> analysed the economic value of Mission Grass-derived carbon materials in SCs. The major results showed that SCs assembled with this BDCM exhibited a low selling price of \$2.37 per unit, lower than those of comparable products on the market, indicating strong market competitiveness.

## Highlight

From life-cycle perspective, the application of BDCMs in the SSC field effectively reduces production costs and improves application benefits. With technological advancements and continuous market expansions, BDCMs will become an integral part of the SSC material sector, contributing to the green transition and sustainable development of the electronics industry and energy storage devices.

## 5. Conclusions and future outlooks

### 5.1 Conclusions

The application of BDCMs in the field of SSCs has reached unprecedented technical heights. BDCMs, with their unique renewability and environmental friendliness, exhibit high power density and exceptional cycling stability when used as electrode materials in SSCs and are leading a revolution in energy storage technology. Sustainably transforming biomass into carbon materials reduces production costs of electrode synthesis and aligns with the trend towards green development from a life-cycle perspective. The significant advancements in this field are closely tied to the collaboration with advanced technologies such as ML and 3D printing. ML empowers new material development and accelerates deep optimization and efficient screening for BDCMs with high-performance electrochemical applications. The 3D printing technical routes facilitate the structural optimization and manufacturing process of BDCMs for SSCs. Moreover, the market for BDCM-based SSCs is expanding at an unprecedented rate, which is driven by the global need for sustainable energy and efficient energy storage solutions. From electric vehicles to wearable devices, smart grids to portable electronics, BDCM-based SSCs are gradually infiltrating and transforming various industries.

With continuous technological advancements and decreasing costs, BDCM-based SSCs are poised to become a significant force in the future energy storage market, driving the green transition and sustainable development of entire energy industry, which is also beneficial to achieving sustainable biomass management and several of UN Sustainable Development Goals. Transforming biomass into carbon-based electrodes for SSCs, in a sustainable and practical manner, strongly facilitates the development and utilization of affordable and clean energy from abundant biomass sources, which is beneficial to achieving UN SDG Goal 7 of affordable clean energy. Secondly, these BDCM-based SSCs, serving as efficient energy storage devices, could be extensively utilized in the construction of sustainable cities and communities, thereby reducing greenhouse gas emissions and enhancing overall urban sustainability and community environments. It suggests that UN SDG Goal 11 of sustainable cities and communities is being promoted and accomplished while applying BDCM-based SSCs for energy supply in our daily-living cities and communities. Thirdly, upcycling biomass into value-added electrode materials is beneficial to achieving a sustainable waste-to-materials strategy and also a circular carbon economy, which is effectively capable of UN SDG Goal 12 of responsible consumption and

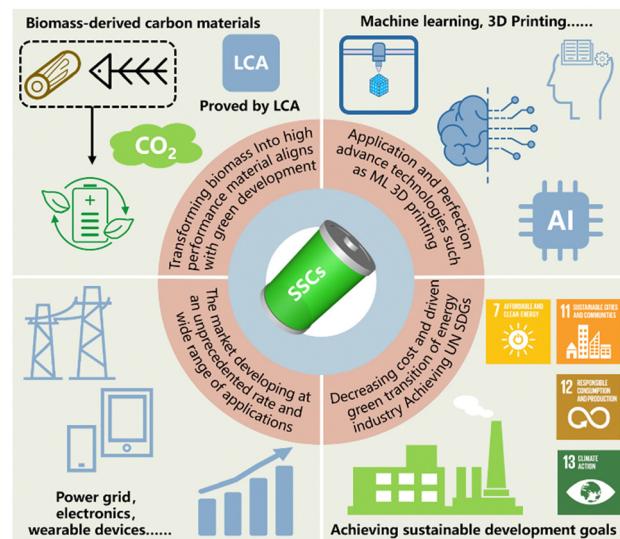


Fig. 7 Sustainably transforming biomass into carbon materials for SSCs achieving energy security solidification and UN Sustainable Development Goals including 7, 11–13.

production. Fourthly, renewable energy-driven BDCM-based SSCs, one carbon-neutral technical route, not only efficiently provide sustainable and steady electricity, but also significantly contribute to climate change mitigation, which is beneficial to achieving UN SDG Goal 13 of climate change and its impacts. With our concerted efforts, sustainably transforming biomass into value-added electrode materials for SSCs plays a significant role in achieving the above-mentioned four UN SDGs. As summarised in Fig. 7, BDCM-based SSCs have not only made breakthroughs in application scope and market growth, but also laid the foundation for the future green transformation of energy and the realization of SDGs.

### 5.2 Future outlooks

SSCs have been widely considered as one of the most promising and feasible energy storage devices. SSCs provide stable power support for battlefield communication equipment under high temperature, low temperature, strong vibration, or other extreme conditions. Due to their high-power density and fast discharge capacity, SSCs improve the fire speed and power of electromagnetic weapons. SSCs meet the energy requirements for unmanned aerial vehicles (UAVs) and unmanned combat vehicles (UCVs) under harsh conditions, achieve rapid take-off and high-speed manoeuvring of UAVs, and are used for energy recovery in UCVs to improve endurance. And SSCs play an important role in battlefield communication, unmanned equipment, electromagnetic launch and so on, and have become an important part of modern military equipment. However, there are still several unresolved issues that limit their commercial applications. Therefore, concerted efforts need to be made in the following aspects:

(1) Applications of cutting-edge technologies for accelerating commercial-scale development of SSCs: as typical advanced approaches, ML and 3D printing play significant roles in

accelerating the development of BDCMs with excellent electrochemical performance for application in SSCs. Compared to the conventional trial-and-error approach, ML exhibits powerful knowledge for accurately designing electrode materials with desired characteristics and revealing relationships among synthesis parameters, textural properties, and electrochemical characteristics of BDCMs in SSCs. The transformation mechanisms need to be illustrated based on experimental results and ML algorithms. Moreover, following the guidelines of ML, 3D printing technologies provide advanced sample preparation routes for transforming biomass into electrode materials in an efficient manner. However, the integration ML with 3D printing is facing huge challenges, such as unavailable databases and guidelines for transforming printed biomass-based carbon precursors into electrode materials.

(2) Circular economy-inspired biomass transformation for achieving UN SDGs: circular economy-inspired biomass management avoids serious environmental pollution caused by improper biomass waste management and achieves the waste-to-resources strategy. However, biomass collected from different sorts and areas exhibits different characteristics, especially for seasonal and regional food waste, posing a critical challenge for developing unified biomass transformation for producing carbon materials for SSCs. It suggests that the development of transformation guidelines for specific biomass might be one practical method for preparing high-performance electrode materials.

(3) Developing advanced manufacturing technologies of electrode materials for SSCs in line with sustainable biomass management: it is essential to continuously explore the advanced transforming routes of biomass precursors into electrode materials with high electrochemical performance. This directly enhances the energy density and power density of SSCs, meeting the demands of future energy storage. Moreover, the environmental benefits and economic feasibility need to be comprehensively assessed for BDCM-based SSCs, especially in the context of carbon neutrality. Biomass, one abundant, carbon-neutral, and well-distributed carbon precursor, has been verified as one promising and practical material for developing high-performance electrode materials. However, based on lab-scale investigations, scaling up to commercial-scale applications is still challenging. Solid efforts need to be made for the deployment of BDCM-based SSCs.

In sum, SSCs, as emerging and promising energy storage devices, require advancements to overcome transformation mechanisms and commercialization barriers. Solid efforts should prioritize sustainable manufacturing and large-scale production technologies to boost electrochemical performance and minimize environmental impacts, while leveraging cutting-edge technologies like 3D printing and ML to continuously optimize BDCMs. Comprehensive life-cycle assessment of biomass transformation into electrode materials provides key knowledge to academic researchers and governmental policymakers, which are also beneficial to achieving circular carbon economy and UN SDGs including Goals 7, 11, 12, and 13.

## Author contributions

Ruibo Fan: conceptualization, data curation, investigation, visualization, writing – original draft, and writing – review and editing; Xiangzhou Yuan: conceptualization, funding acquisition, project administration, supervision, writing – original draft, and writing – review and editing; and Beicheng Xue, Pengfei Tian, Xuesong Zhang, and Huiyan Zhang: writing – review and editing.

## Data availability

No primary research results, software or code have been included and no new data were generated or analysed as part of this highlight.

## Conflicts of interest

There are no conflicts to declare.

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