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REVIEW

Recent advances in the application of carbon-based electrode materials for high-performance zinc ion capacitors: a mini review

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Abstract
Designing and developing advanced energy storage equipment with excellent energy density, remarkable power density, and outstanding long-cycle performance is an urgent task. Zinc-ion hybrid supercapacitors (ZIHCs) are considered great potential candidates for energy storage systems due to the features of high power density, stable cycling lifespans, high safety, low cost, and long-term durability, which originate from the combination of the dual advantages of supercapacitors and zinc-ion batteries. Recently, to pursue the long lifespan of ZIHCs, effective progress has been made in the development and application of ZIHC cathode materials based on carbon-based materials. This review takes carbon-based materials as the starting point and discusses the charge storage mechanism of ZIHCs. Moreover, the application of various carbon-based materials is systematically summarized in ZIHCs, including activated carbon (AC), biomass carbon (BC), porous carbon (PC), and heteroatom-doped carbon (HDC). In addition, recent advances in the structural design of electrolytes and Zn anodes and their effects on electrochemical performance are summarized. Ultimately, the current challenges and the potential directions for ZIHCs are presented. This paper intends to provide directions for the further development of high-performance ZIHCs.

Keywords Zinc-ion hybrid supercapacitor · Carbon-based electrodes · Electrochemical performance

1 Introduction

The sustainable development goals of modern society have prompted the world to focus on conserving energy resources and implementing a comprehensive conservation strategy [1–7]. The rapid development and utilization of new and recyclable energy sources, including solar energy and wind energy, impels the exploration of energy storage devices, such as batteries and supercapacitors [8–14]. The electrochemical performance of different storage devices generally depends on their energy storage mechanism. The cell undoubtedly has excellent energy storage capacity, with the cation departing from the cathode into the electrolyte and then adsorbing/inserting itself into the anode. During discharge, they are then un-inserted from the anode into the electrolyte and returned to the cathode. At the same time, the anion moves from the electrolyte and adsorbs onto the anode. However, the slow energy storage rate results in high energy density but low power density. Unlike batteries, supercapacitors (especially electric double-layer capacitors) absorb charge at the surface of the electrode material, and the ions in the electrolyte move toward the positive and negative electrodes, respectively, during charging, thus allowing reversible charging and discharging processes at very fast speeds with the high power density and low energy density [15, 16]. Furthermore, the characteristic ultralong cycling performance of supercapacitors is superior to batteries, which is irreplaceable in practical applications that require high power density and ultralong cycle capability, such as airplanes and powered cars [17]. Nevertheless, the practical widely application was hindered based on its low energy density [18–21]. Consequently, multitudinous strategies have been carried out to enhance the energy density based on the advantages of high power density and ultralong cycling stability to broaden its application fields [22–24].

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The energy density for the supercapacitor is calculated via Eq. (1):

$$E = \frac{1}{2}CV^2 \tag{1}$$

where E represents the energy density, C signifies the specific capacitance of the active material, and V means the working voltage. Obviously, the energy density of supercapacitors can be enhanced mainly according to the improvement of the capacity of the active material and the operating voltage. In addition to optimizing electrolytes, designing and building special structures of capacitor devices, especially combining the advantages of capacitor-type electrodes with battery-type electrodes, is believed to be effective to improve its working voltage for obtaining the ideal performance of supercapacitors [25–28].

Among the multitudinous research, metallic Zn has been chosen as an excellent anode electrode for supercapacitors systems due to its superiority of low standard potential, high security, low cost, environmental protection, and mass production. The zinc-ion hybrid supercapacitors (ZIHCs) consist of the superiority of supercapacitors and Zn-ion batteries, with Zn sheets as negative electrodes to broaden the operating voltage, which was believed to have immense potential in improving the low energy density of conventional supercapacitors [29–31]. The concept of ZIHCs with carbon nanotubes and metallic Zn as electrode materials was creatively put forward in 2016, which delivered a capacitance of 53 F g^{-1} [32], setting off a research boom in ZIHCs. A high capacitance of 436 F g^{-1} and long-cycle stability of ZIHCs using carbon-based electrode materials was achieved in a recent study [33], which shows a bright, practical application prospect of ZIHCs in terms of carbon-based cathodes.

Besides numerous explorations on Zn-anode protection, enormous attention has been paid to carbon-based cathode materials that store charge through electrochemical double layers (EDLC) and pseudo-capacitance effects [34]. EDLC capacitance relies on their rich specific surface area and pore structure that allows extensive active site contact between electrolyte ions and the carbon cathode, while the pseudo-capacitance capacitance is related to the surface functional groups of the cathode material or the occurrence of redox reactions. Therefore, the design of carbon-based nanostructures with large specific surface area or/and by introducing heteroatoms [35], conducting polymers [36], organic molecules [37], and metal oxides [38] are essential for the enhancement of EDLC and pseudo-capacitance property.

In this review, the research progress and challenges of ZIHCs were presented, which can be summarized in Fig. 1. The principle of ZIHC, mainly including the energy storage mechanism and energy calculation method, is first introduced. Then, the research progress of ZIHCs with carbon-based cathode materials is comprehensively described, such as the improvement project of activated carbon, biomass carbon, and other materials. Besides, the effects of electrode structure design and emerging electrolyte systems on the electrochemical performance of ZIHCs are reviewed. Finally, the technological challenges and the future development direction of ZIHCs are discussed. This review will offer a more profound perception of carbon-based electrodes for ZIHCs and assists with technological innovation.

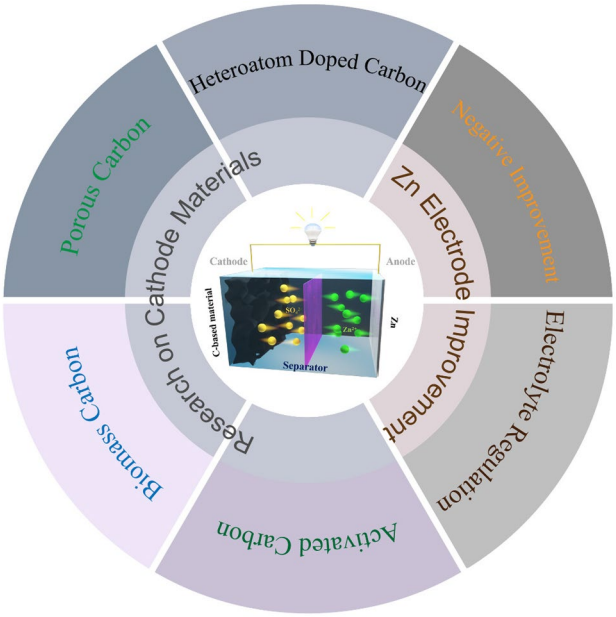

Fig. 1 Summary of the aspects discussed in this review

2 Energy storage mechanisms of ZIHCs

Theoretically, the capacity of ZIHCs is considered to have no upper limit due to the negative zinc electrode exhibiting Faraday capacitance and a flat potential platform. Therefore, the capacitance of ZIHCs almost entirely originates from the contribution of carbon materials (Eq. (2)); meanwhile, ZIHCs present better capacitance behaviors than symmetric supercapacitors [39]. However, some other voices and arguments have emerged in view of the unsatisfactory capacitance properties certified by some recent anion adsorption/desorption experiments [40, 41]. On the one hand, some researchers stick to the view that the adsorption/desorption of Zn^{2+} on carbon-based composites contributes to the capacitance (Eq. (3)) [42, 43]. On the other hand, the capacitance is believed to be increased by the adsorption/desorption of anions on carbon-based hybrids (Eq. (4)) [44]. For

example, three different zinc cation electrolytes (zinc sulfate, zinc acetate, and zinc chloride) were applied to probe the influence of electrolyte anions on the electrochemical properties of zinc-ion hybrid capacitors, demonstrating the role of anions in electrochemical energy storage [45]. In summary, the future development of supercapacitors is injected with new energy in the emerging ZIHCs devices while brightening the route to achieving capacitors with better electrochemical properties. The complex energy storage principle of ZIHCs has been gradually established with constant explorations.

$$C = \frac{1}{\frac{1}{C_+} + \frac{1}{C_-}} \tag{2}$$

$$C + X^- \leftrightarrow C||X^- \tag{3}$$

$$C + Zn^{2+} \leftrightarrow C||Zn^{2+} \tag{4}$$

ZIHCs are electrochemical devices consisting of battery-type anodes and capacitor-type cathodes; hybrid structures consist of zinc-ion batteries and supercapacitors (Fig. 2) [46]. During the charging process, the anions in the electrolyte move and adsorb to the cathode (or are trapped in the cathode) in order to obtain an electric double layer (EDLC), while the Zn^{2+} in the electrolyte moves and deposits on the zinc anodes. The discharge process is the opposite of the charging process, with the anions and Zn^{2+} on the electrode diffusing back into the electrolyte. The cathode electrodes of ZIHCs always adopt the carbon-based structure with large porosity to accommodate the intercalation/extraction of ions, which are separated from the metallic zinc anodes by a permeable insulating separator. And soluble zinc salt solutions are usually applied as electrolytes in ZIHCs due to the high stability, low viscosity, and high ion transport rate of Zn^{2+} in aqueous solutions [47]. However, aqueous electrolytes always lead to a limited voltage range between 0.2 and 1.8 V based on the competitive decomposition of water. The non-aqueous electrolytes were reported to increase the voltage range to exceed 1.8 V but with problems of safety and economy [48]. In contrast, electrolyte improvement is a superior measure, which will be discussed in "Performance regulation of ZIHCs."

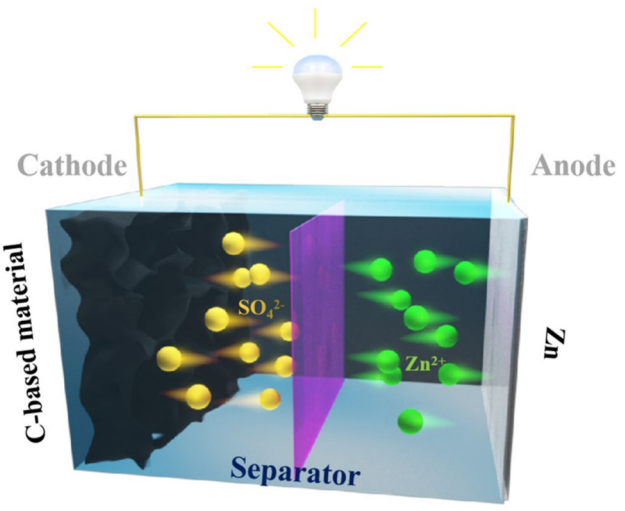

Fig. 2 Schematic representation of ZIHC composed of carbon-based cathodes and zinc anodes

3 Carbon-based electrode materials for ZIHCs

Carbon-based materials have been tremendously investigated as supercapacitor electrodes in terms of their high hole density, large specific surface area, outstanding conductivity, excellent stability, abundant raw materials, and safety [49–52]. In previous studies on ZIHCs, the charge storage capacity of carbon materials was demonstrated to be originated from the interface of the electrode/the electrolyte, and the electrochemical performance of ZIHCs was also influenced by the mentioned factors. Therefore, multitudinous carbon materials, including biomass carbon (BC), activated carbon (AC), heteroatom-doped carbon (HDC), and porous carbon (PC), are explored as electrodes for ZIHCs, and satisfactory performance was obtained. The recent works on ZIHCs consisting of carbon-based and Zn electrodes with ideal electrochemical properties are listed in Table 1, which illustrates their promising practical commercial application.

3.1 Activated carbon material

Activated carbon materials have been widely investigated as potential electrode materials for ZIHCs, which exhibit excellent characteristics of large specific surface area, easily tunable porosity, low consumption, and high stability [65]. For instance, a zinc-ion hybrid capacitor consisting of commercial activated carbon (AC) as the cathode, metallic Zn as anodes, and Zn sulfate aqueous solution as the electrolyte was constructed by Dong et al. (Fig. 3a). The commercial AC displayed a large specific surface area of 1923 $m^2\ g^{-1}$ due to its irregular surface particles and the rough morphology (Fig. 3b, c). The specially designed ZIHCs exhibited stable CV performance at scan rates of 2–1000 mv s^{-1} (Fig. 3d, e), presented a capacitance of 121 mAh g^{-1} and an energy density of 84 Wh kg^{-1} at 0.1 A g^{-1} (Fig. 3f, g), a high power output of 14.9 KW kg^{-1}, and finally high capacity retention over 91% (Fig. 3i) that was achieved after 10,000 cycles [26]. For the sake of further investigating the effects

Table 1 Electrochemical characteristics summary of carbon-based materials in ZIHCs

Materials	Electrolyte	Voltage range (V)	Capacity	Current density (A g^{-1})	Electrochemical performance	Energy density (W h kg^{-1})	Power density (kW kg^{-1})	Ref.
AC	1 M $Zn(CF_3SO_3)_2$	0–1.7	170 F g^{-1}	0.1	91% after 20,000 cycles	52.7	1.725	[53]
AC	2 M $ZnSO_4$	0.2–1.8	121 mA h g^{-1}	0.1	91% after 10,000 cycles	84	14.9	[26]
AC	2 M $ZnSO_4$	0–1.8	72.1 mA h g^{-1}	0.05	100% after 10,000 cycles	115.4 μWh cm^{-2}	3.9 mW cm^{-2}	[54]
CNT	1 M $ZnSO_4$	0.2–1.8	83.2 mF cm^{-2}	1 mA cm^{-2}	87.4% after 6000 cycles	29.6 μWh cm^{-2}	0.8 mW cm^{-2}	[41]
PSC-A600	1 M $Zn(CF_3SO_3)_2$	0.2–1.8	183.7 mA h g^{-1}	0.2	92.2% after 10,000 cycles	147	0.136	[44]
MSAC	2 M $ZnSO_4$	0.3–1.8	176 mA h g^{-1}	0.5	78% after 40,000 cycles	188	0.533	[55]
HPC	2 M $ZnSO_4$ @1 M Na_2SO_4	0.01–1.8	204 mA h g^{-1}	0.2	94.9% after 2000 cycles	118	0.2	[56]
PCN	1 M $ZnSO_4$	0.1–1.7	149 mA h g^{-1}	0.2	91% after 10,000 cycles	119	0.16	[57]
PCNFs	1 M $ZnSO_4$	0.1–1.7	177.7 mA h g^{-1}	0.5	90% after 10,000 cycles	142.2	0.4	[58]
MPC	1 M $Zn(CF_3SO_3)_2$	0–1.8	209 F g^{-1}	0.2	100% after 10,000 cycles	92.7	0.179	[59]
HCSs	2 M $ZnSO_4$	0.15–1.95	86.8 mA h g^{-1}	0.5	98% after 15,000 cycles	59.7	0.448	[60]
B/N co-doped LDC	1 M $ZnSO_4$	0.2–1.8	127.7 mA h g^{-1}	0.5	/	97.6	12.1	[40]
P@B-AC	2 M $ZnSO_4$	0.2–1.8	169.4 mA h g^{-1}	0.5	88% after 30,000 cycles	169.4	20	[61]
N-HPC	2 M $ZnSO_4$	0.2–1.8	136.8 mA h g^{-1}	0.5	90.9% after 5000 cycles	191	3.633	[62]
B-diamond/ carbon fiber	1 M $ZnSO_4$	0.2–1.8	35.1 F g^{-1}	0.2	89.9% after 10,000 cycles	70.7	0.709	[63]
N-PC	2 M $ZnSO_4$	0.15–1.7	302 mA h g^{-1}	1.0	> 100% after 10,000 cycles	157.6	0.69	[64]

of specific surface area on AC electrodes for ZIHCs, a high-performance ZIHC was assembled by a three-dimensional porous AC material prepared by an organic salt precursor as the cathode with a high specific surface area of 2854.1 m^2 g^{-1} and a rich micro/ultra-microporous structure, which exhibited a capacitance of 213 mAh g^{-1}, energy density of 164 Wh kg^{-1} and the power density of 390 W kg^{-1} at t 0.5 A g^{-1}, and high capacity retention of 90% after 20,000 cycles at 10.0 A g^{-1} [66].

In short, the application of activated carbon materials ZIHCs is an innovative attempt and breakthrough; the feasibility of carbon materials on ZIHCs has been demonstrated based on the successful experimental results, which will lead the way for the subsequent application of other carbon material electrodes in ZIHCs.

3.2 Biomass-derived carbon material

Recently, the reuse value of various raw material-rich and renewable biomass wastes has been favored by researchers [67]. Biomass-derived carbon (BC) materials have the advantages of large specific surface area, tunable microstructure, and surface functional groups, which gradually become the most promising candidate materials for electrodes under the promotion of the two-carbon policy. For example, biomass-derived carbon stem from coconut shells was synthesized with a specific surface area of 3554 m^2 g^{-1} and was applied as electrodes for ZIHCs (Fig. 4a–c), which presented a capacity of 170 F g^{-1} at 0.1 A g^{-1} (Fig. 4d–f) and an energy density of 52.7 Wh kg^{-1} at 1725 W kg^{-1} that calculated based on the active substance. Besides, the capacity retention can be up to

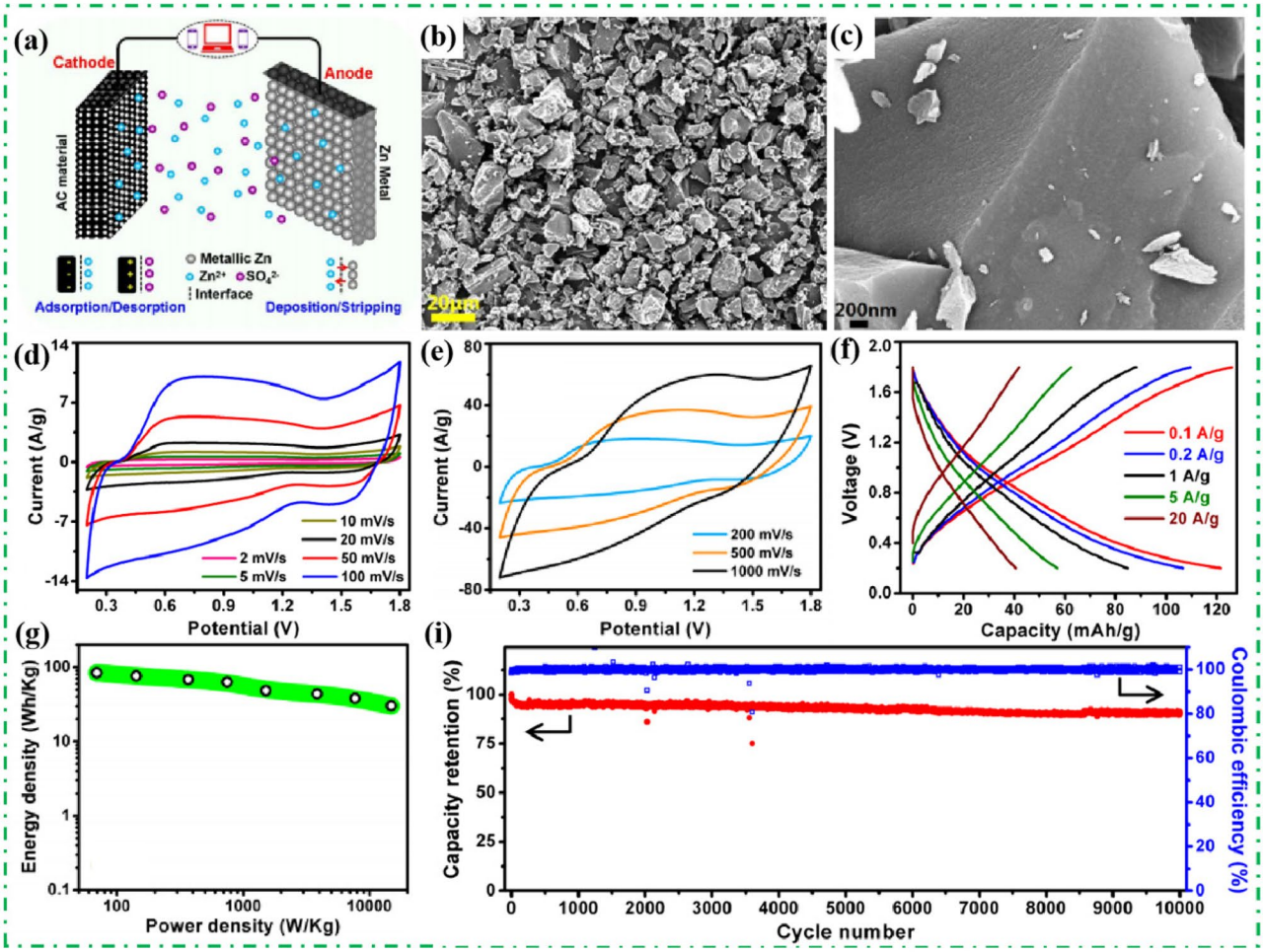

Fig. 3 Morphology and electrochemical properties of AC: **a** schematic of AC//$ZnSO_4$ (aq)//Zn system; **b**, **c** SEM images of AC powder; **d**, **e** CV curves at 2.0–1000 mV s^{-1}; **f** GCD curves at 0.1–20 A g^{-1}; **g** Ragone plot; **h** cycling performance at 1.0 A g^{-1} of ZIHCs. Reproduced with permission from [26], Copyright 2018, Elsevier

nearly 100% after 22,000 lifespans at 1.0 A g^{-1} (Fig. 4g) and over 91% after 20,000 cycles even at 2.0 A g^{-1} (Fig. 4h) [53]. Meanwhile, corncob-derived carbon materials were prepared by single-step calcination of the low-cost corncob wastes, which were applied as cathode electrodes for ZIHC (Fig. 5a). The specific surface area of active corncob-derived carbon (ACC) materials reached up to 2619 m^2 g^{-1} after the single-step carbonization and activation (Fig. 5b,c) and exhibited exceptional specific capacitance in an alkaline electrolyte (Fig. 5d), acidic electrolyte (Fig. 5e), and neutral electrolyte (Fig. 5f). Simultaneously, the GCD curves of the ZIHC presented the typical linear shapes (Fig. 5g), an energy density of 94 Wh kg^{-1} at 68 W kg^{-1} (Fig. 5h), and good cycling stability of high capacity retention over 98.2% after 10,000 lifespans at 5.0 A g^{-1} (Fig. 5i) [68]. Furthermore, a high mesoporous carbon material originating from agricultural waste corn silk was prepared and showed excellent energy storage performance for both supercapacitor and ZIHCs. The assembled supercapacitor delivered an energy density of 25 Wh kg^{-1} and a remarkable cycling lifespan at a high power density of 23,070 W kg^{-1}; in contrast, a high energy density of 38.5 Wh kg^{-1} at the high power density of 14,741 W kg^{-1} for the ZIHC was achieved. The results not only demonstrated the feasibility of mesoporous carbon materials as electrode materials for supercapacitors and ZIHCs but also reflected the superior electrochemical performance of ZIHCs compared with supercapacitors [69].

Compared with activated carbon, biomass-derived carbon materials present the advantages of an abundant resource and adjustable surface chemical information and are beneficial to the sustainable utilization of resources and the establishment of an environment-friendly society.

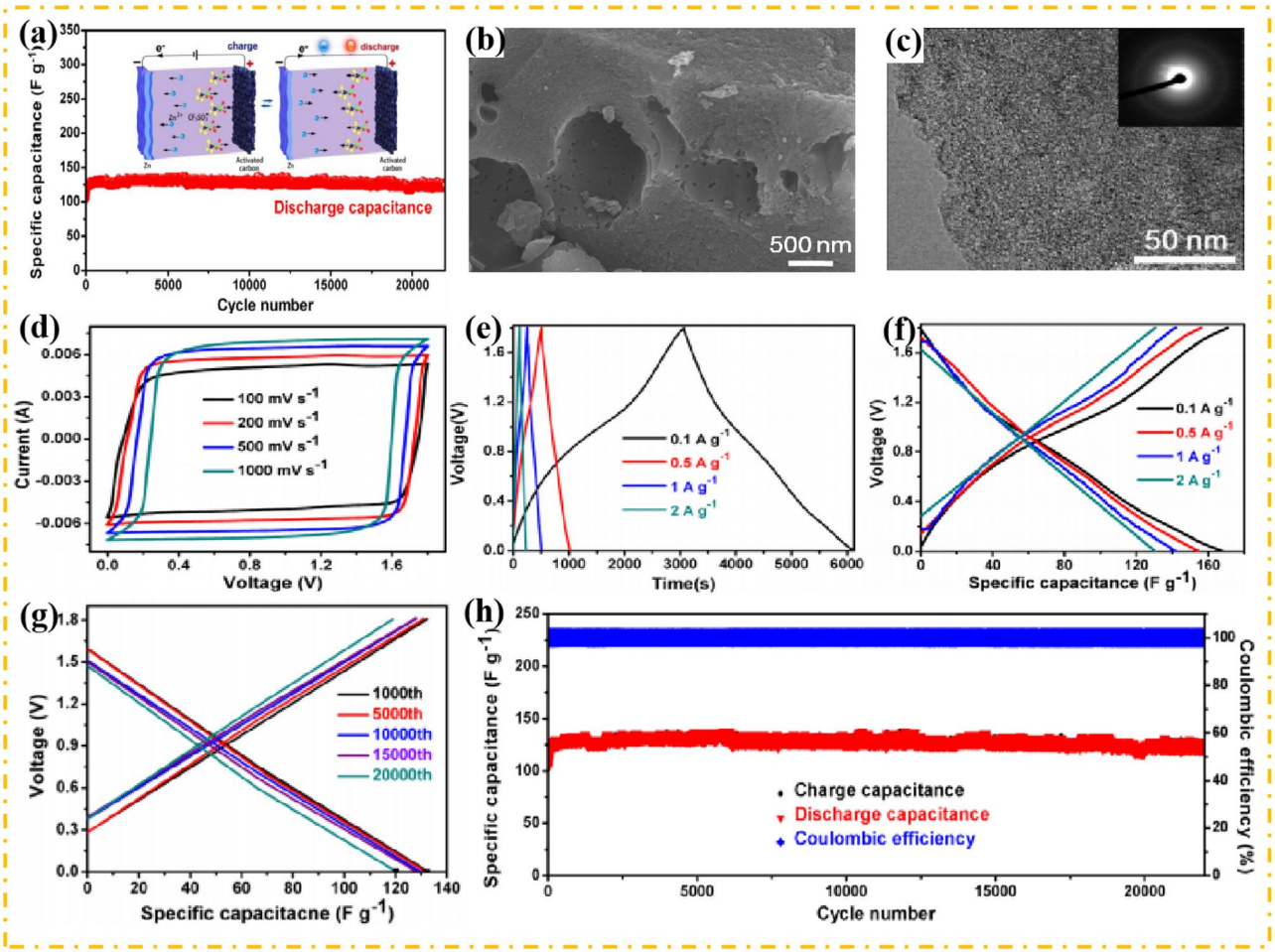

Fig. 4 Morphology and electrochemical properties of BC derived from coconut shells: **a** schematic diagram of ZIHC working principle and cycle performance. **b** SEM and **c** TEM image of coconut shell. **d** CV curves of ZIHC at 100–1000 mV^{-1}. (**e**) GCD curves of ZIHC at 0.1–2.0 A g^{-1}. **f** Voltage-specific capacitance curves of ZIHC at 0.1–2.0 A g^{-1}. **g** GCD curves of the ZIHC after 1000–20,000 cycles. **h** Cycling property of the ZIHC at 2.0 A g^{-1}. Reproduced with permission from [53], Copyright 2017, Elsevier

3.3 Porous carbon material

The significant characteristic of porous carbon materials roots in the abundant pore structure that enables the regulation of specific surface area. In addition, the advantages of abundant raw materials and easy regulation promote the wide explorations of porous carbon acting as cathode electrodes for ZIHCs [70]. Two-dimensional porous carbon nanoflates (PCNF) were synthesized via chemical etching technique using sodium polyacrylate powder mixed with potassium bicarbonate powder [58]; the preparation routes are shown in Fig. 6a. The as-prepared PCNF presents submitted a stacked nanosheet-like structure (Fig. 6b), and the pore structure is identified by the HRTEM image (Fig. 6c); the specific surface area of PCNF-4 is as high as 1770 m^2/g, which presented satisfactory capacity retention over 90% after 10,000 lifespans at 10.0 A g^{-1} (Fig. 6d). The ZIHC assembled with PCNF as cathode achieved an energy density of 142.2 Wh kg^{-1} at a power density of 400.3 W kg^{-1} at 0.5 A g^{-1}, an energy density of 68.4 Wh kg^{-1} even at the power density of 15,390 W kg^{-1} at 20 A g^{-1} (Fig. 6e), which demonstrated the excellent energy output capability of ZIHCs. Furthermore, two green diodes were lighted by the assembled ZIHCs, which demonstrate their potential application (Fig. 6f). Meanwhile, a graded porous carbon (PSC) stem from pencil shavings with hierarchical porous structure and high specific surface area of 1293 m^2/g was prepared (Fig. 7a) [43]. The ZIHC consisted of PSCs as positive and metallic Zn as negative electrode companied with $Zn(CF_3SO_3)_2$ solution as electrolyte exhibited superior rate capability and ultrahigh exceptional capacity of 183.7 mA g^{-1} (Fig. 7b–d), a maximum energy of 147.0 kW kg^{-1}, a maximum power density of 15.7 kW kg^{-1} (Fig. 7e), and an impressive 92.2% capacitance retention after 10,000 lifespans at 10.0 A g^{-1} (Fig. 7f). More surprisingly, the ZIHC maintained

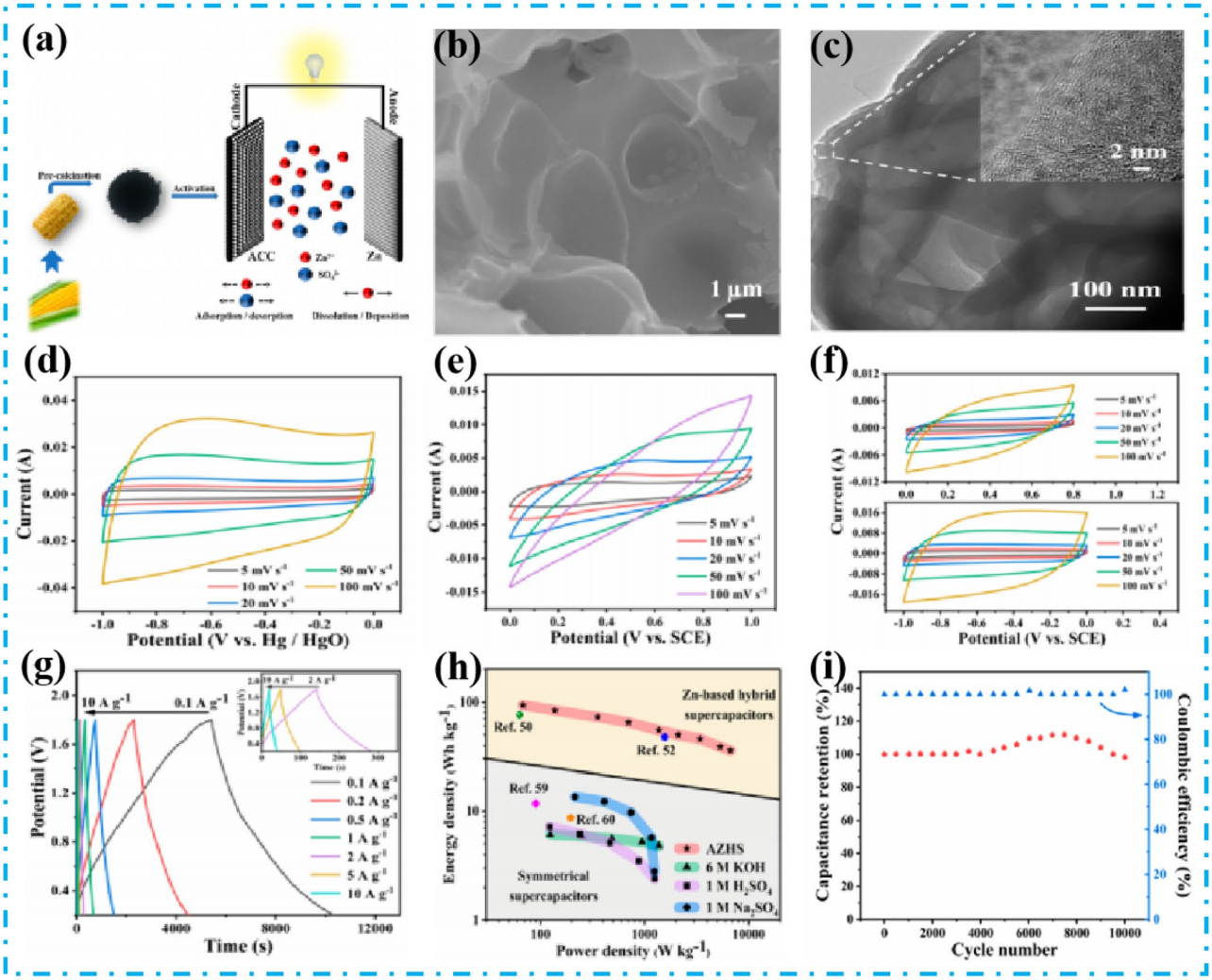

Fig. 5 **a** Diagrammatic sketch of corncob applied as an electrode for ZIHC. **b** SEM image and **c** TEM image of ACC. Three-electrode electrochemical performance of ACC samples in **d** KOH electrolytes, **e** H_2SO_4 electrolytes, and **f** Na_2SO_4 electrolytes. **g** GCD curves of ZIHC at 0.1–10.0 A g^{-1}. **h** Ragone plots of ZIHC and some supercapacitors reported in the literature. **i** Cycling performance of ZIHC at 5 A g^{-1}. Reproduced with permission from [68], Copyright 2019, ACS

excellent antifreeze performance (nearly 100% coulombic efficiency at − 15 °C) and cycling stability (over 80 cycles) when antifreeze hydrogel is added to the electrolyte system (Fig. 7g). Besides, a 3D hierarchical porous carbon (3D HPC) derived from waste bagasse, and coconut shells was obtained by process of hydrothermal carbonization and potassium hydroxide activation (Fig. 8a) [56]; the strong porosity gives HPC a high surface area of 3401 m^2 g^{-1} and a large pore volume of 2.03 cm^3 g^{-1}. The porous structure (Fig. 8b–d) endows it with good electronic conductivity, resulting in excellent electrochemical performance when assembled into ZIHCs as a cathode. ZIHCs exhibit a capacitance of 305 mAh g^{-1} at 0.1 A g^{-1} (Fig. 8e), a high energy density of 118 Wh kg^{-1}, outstanding rate performance, and remarkable cycling stability with a capacitance retention rate of 94.9% after 20,000 cycles at 2.0 A g^{-1} (Fig. 8f).

The porous carbon materials endowed with excellent adsorption properties are believed to be one of the most promising and popular carbon-based composites in the future. The rational synthesis of porous carbon materials through the targeted design of reaction routes, surface modification, etc. is of outstanding significance for enhancing the capacity of ZIHCs.

3.4 Heteroatom-doped carbon

Compared with pure carbon materials, the introduction of heteroatoms into the carbon matrix is regarded as an effective way to enhance their electrochemical properties. Among

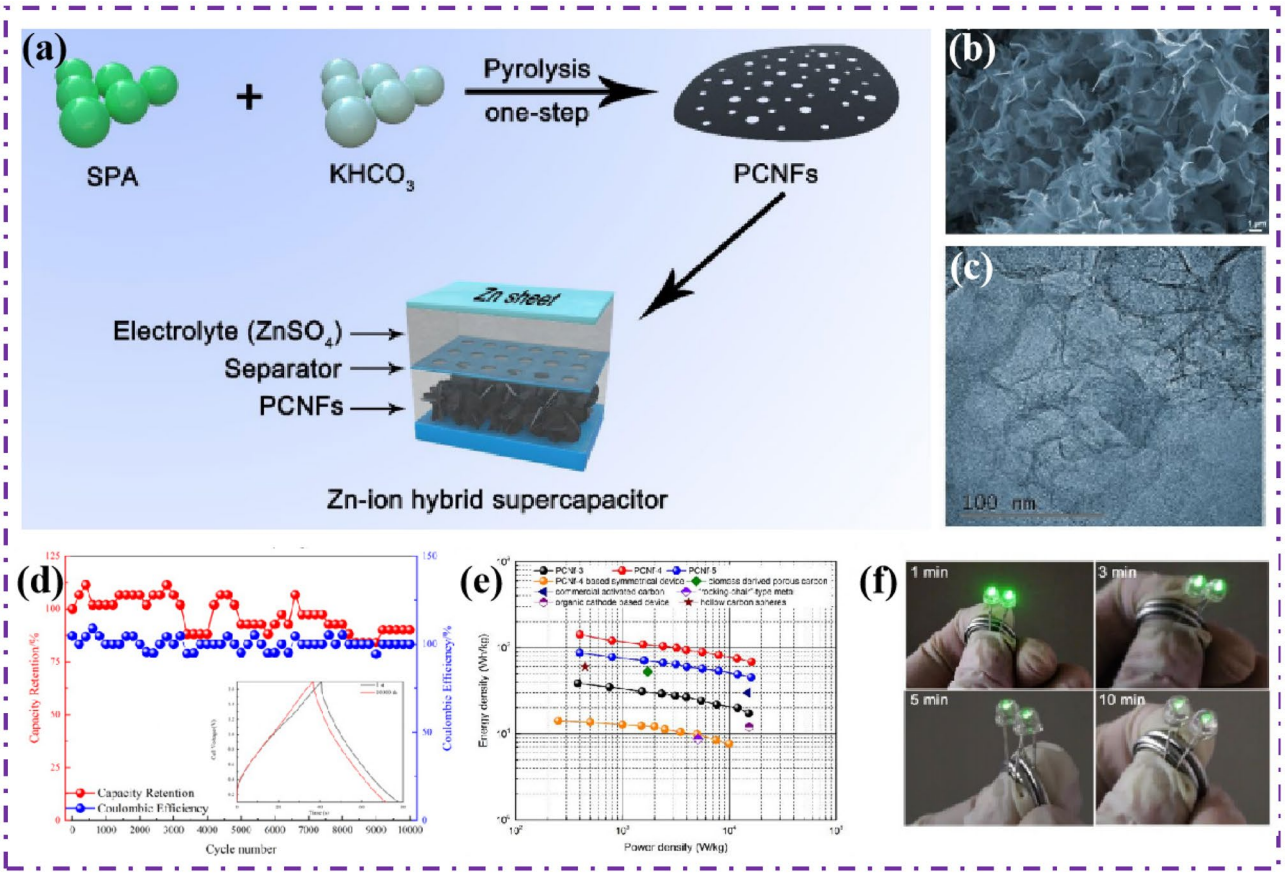

Fig. 6 Morphology and electrochemical properties of PCNF: **a** schematic diagram of the synthesis process and energy storage application of PCNFs. (**b**) FESEM images of PCNF-4. **c** TEM images of PCNF-4. **d** Cycling performance of ZIHC at 10 A g^{-1} (insert is the GCD curves). **e** Ragone plots of PCNFs-based ZIHCs, PCNF-4-based symmetric supercapacitors, and other previously reported ZIHCs. **f** Photograph of two PCNF-4-based ZIHCs lit up with two green LEDs after charging for 60 s at 5 A g^{-1}. Reproduced with permission from [58], Copyright 2020, Elsevier

various heteroatoms, nitrogen atom with large electronegativity contributes to the improvement of electronic conductivity and active site numbers, promoting the increase of capacitance, rate capability, and cycling stability, which is the most widely investigated and promising heteroatom dopant [71]. For example, an aqueous ZIHC consisting of a layered N-doped carbon cathode and metallic Zn anode with high energy and cycling stability was proposed [72]. The ZIHC devices exhibited a high energy density of 107.3 Wh kg^{-1}, an excellent power density of 24.9 kW kg^{-1}, and satisfactory capacity retention beyond 99.7% after 20,000 cycles at 4.2 A g^{-1}, which proved the chemisorption process of Zn^{+}, surface wettability, and conductivity of the electrode material are enhanced by the introduced nitrogen heteroatoms. In addition, a nitrogen and oxygen co-doped-layered HPC/CC synthesized by drop coating and carbonization was reported by Zhao et al. (Fig. 9a) [73]. The uniquely designed composites with plentiful nitrogen and oxygen dopants also presented large pore volumes due to the layered pore construction of micro- and middle holes (Fig. 9b) and exhibited excellent electrochemical behaviors as ZIHC cathodes, including a high capacity of 138.5 mAh g^{-1} at 0.5 A g^{-1}, superior rate capability at 20.0 A g^{-1}, and superior Zn^{+} storage performance (Fig. 9c). Most notably, the original capacity increased to 104.3% after 10,000 cycles because the pores were slowly penetrated by electrolyte ions during cycling (Fig. 9d).

Besides nitrogen and oxygen heteroatoms, other heteroatoms can be applied to dope into the carbon matrix to enhance the storage behaviors of Zn^{+}. For instance, the wettability of electrolyte/electrode can be improved by doping phosphorus elements. Meanwhile, more structural defects would be generated due to the different van der Waals radii of phosphorus (195 pm) and carbon (180 pm). The boron elements present a similar van der Waals radius and valence electron number of carbon, which hardly affects the lattice parameters of carbon but can improve the electronic conductivity [74]. A boron and phosphorus co-doped-activated carbon material (P, B-AC) was achieved by a one-step doping calcination method (Fig. 10a–c), which displayed a high

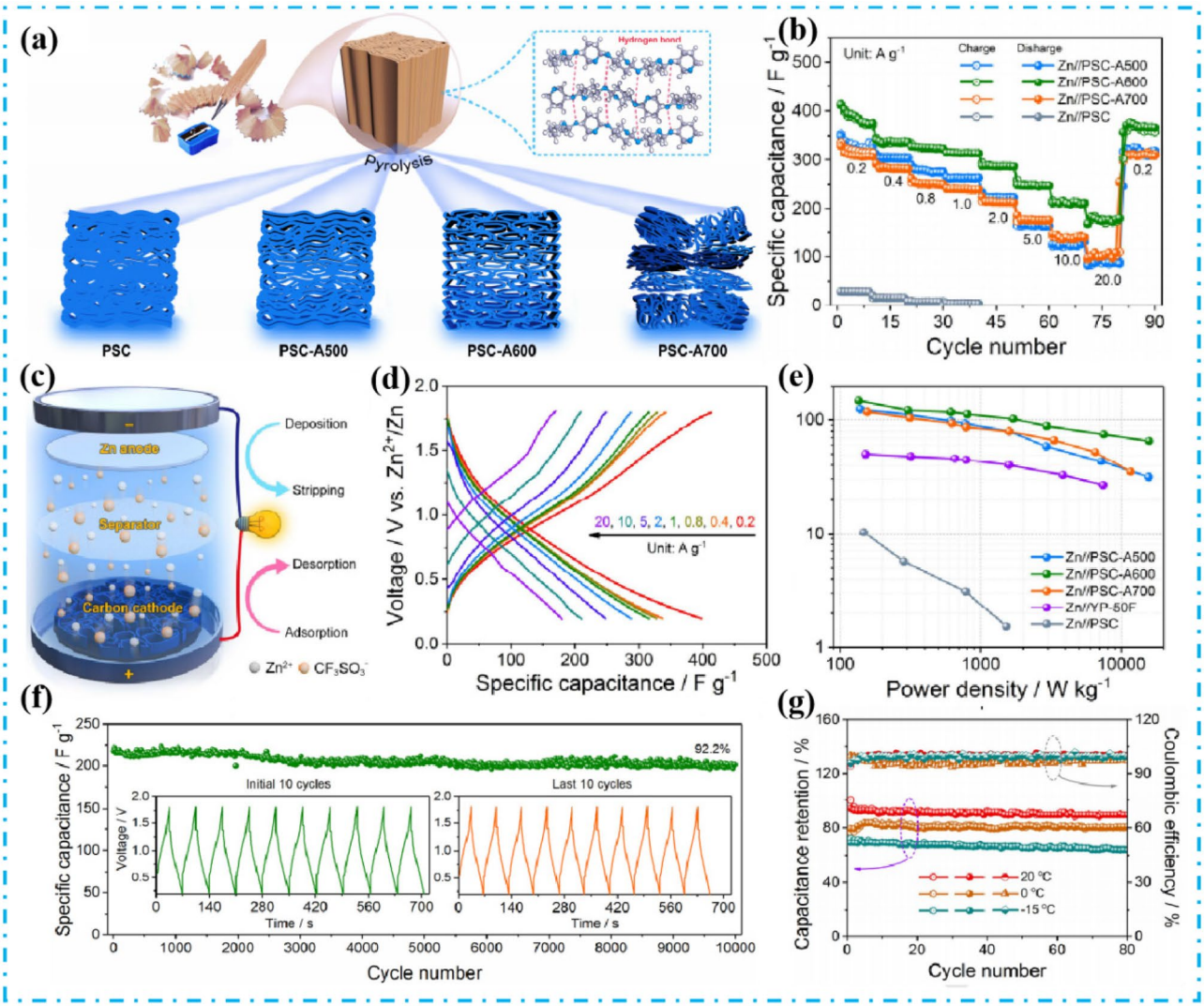

Fig. 7 Morphology and electrochemical properties of PSC: **a** schematic diagram of PSCs. **b** Rate capability of PSCs. **c** Schematic diagram of the structure of ZIHC. **d** GCD profiles of the PSC-A600 at 0.2 ~ 20.0 A g^{-1}. **e** Ragone plots of PSCs. **f** Cycle graph over 10,000 times (insert is the first and last 10 GCD curves). **g** Capacity retention at low temperatures. Reproduced with permission from [44], Copyright 2020, Elsevier

capacitance of 169.4 mAh g^{-1} at 0.5 A g^{-1} (Fig. 10d, e), superior ion mobility (Fig. 10f), and an energy density of 169.4 Wh kg^{-1} and exhibited an excellent power density of 500 W kg^{-1} and capacity retention over 88% after 30,000 cycles (Fig. 10g).

In general, it is a promising strategy to construct carbon matrix cathodes for high-performance ZIHCs by introducing various heteroatoms in terms of their distinctive electronic structures and properties. However, precisely tuning the morphological structure of the doping and achieving stable reproducibility is still a big challenge. Furthermore, the mechanism of action of the synergistic effect of different heteroatom doping is also one of the directions that need to be further explored.

In short, carbon-based materials are generally characterized by a large surface area to allow fast and easy access to the electrode surface, absorbing electrolyte ions to the surface of the electrode material and thus improving the electrochemical performance.

Although enormous research has been focused on zinc ion supercapacitor carbon-based material electrodes, their practical applications are still limited, and advanced carbon-based electrodes are still in great demand.

4 Performance regulation of ZIHCs

The short circuits and cell failure of ZIHCs would occur due to the puncture of the electrolyte diaphragm caused by the formation and growth of zinc dendrite. Furthermore, more

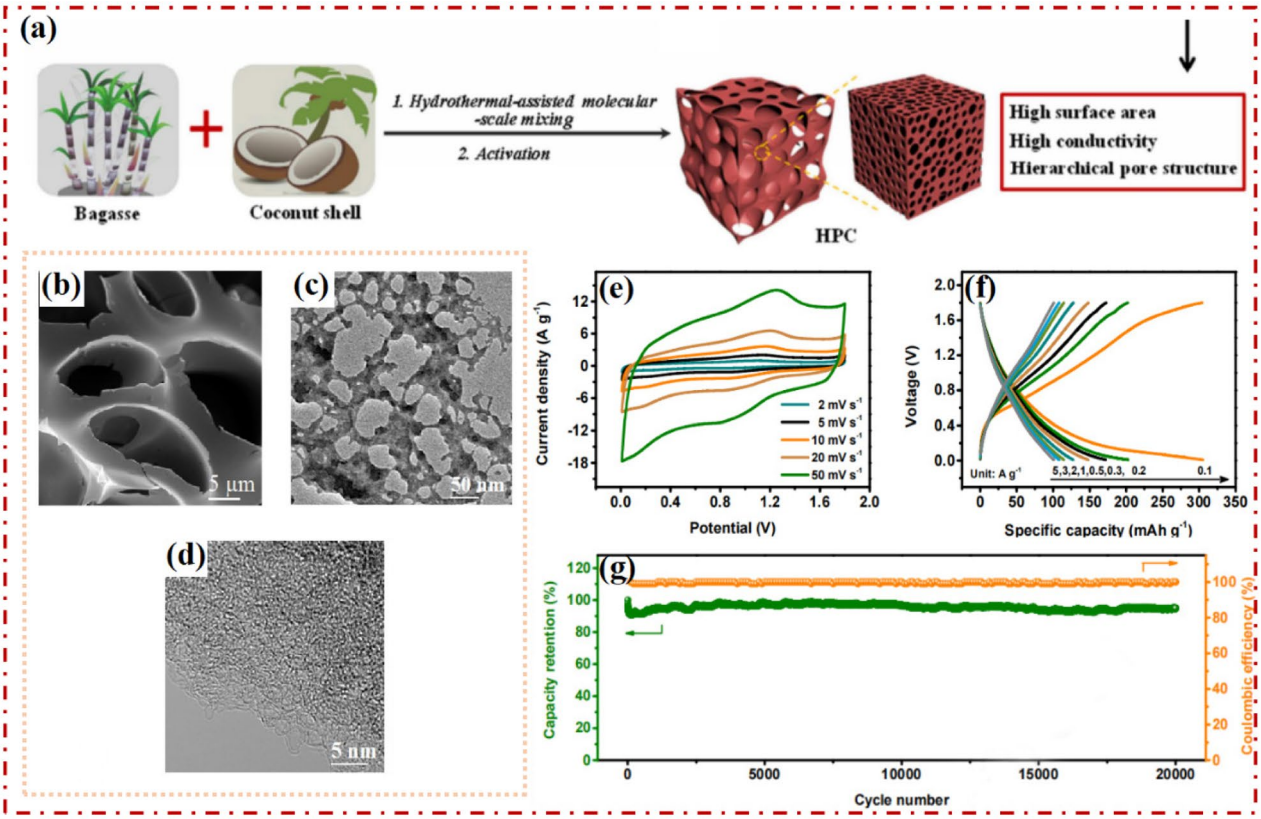

Fig. 8 Morphology and electrochemical properties of HPC: **a** schematic diagram of HPC. **b** SEM image, **c** TEM image, and **d** HRTEM of HPC. **e** CV curves of HPC-based ZIHC at 2.0–50.0 mV s^{-1}. **f** GCD profiles of HPC-based ZIHC at 0.1–5.0 A g^{-1}. **g** Cycling stability at 2.0 A g^{-1} over 20,000 cycles of HPC-based ZIHC. Reproduced with permission from [56], Copyright 2019, Elsevier

severe dendrite occurs in alkaline electrolytes compared with neutral or essential electrolytes, which may be attributed to the $Zn(OH)_4^{2-}$ produced by oxidation or self-corrosion at Zn anode in alkaline electrolytes [75]. The formed $Zn(OH)_4^{2-}$ unevenly distributes on the surface of the zinc electrodes and accumulates gradually and prominently, and uneven Zn negative surface current accelerates the formation of Zinc dendrites, leading to the failure of electrochemical behaviors [76]. The obvious one-dimensional (1D) zinc dendrites are always observed even at low current densities in alkaline electrolytes, but hardly observed at low current densities in neutral and acidic electrolytes [77], which promote the more widespread application of acid or neutral electrolytes for ZIHCs. Nevertheless, obtaining ZIHCs with desirable cycling performance in acid/neutral electrolytes is challenging because numerous large-sized dendrites are still generated, aggregated, and nucleated on the Zn anode at high current densities, causing the performance paralysis of the capacitor devices. Consequently, electrode surface modification and the introduction of additives into electrolytes are effective strategies to alleviate the formation of zinc dendrites and improve the electrochemical performance of ZIHCs, which are becoming one of the focuses in the further exploration of ZIHCs.

4.1 Improving the negative electrodes

Currently, the introduction of coatings on the surface of Zn electrode and/or designing unique structured zinc anodes are regarded to be the most direct, effective, and extensive research strategies for restraining the formation and growth of zinc dendrite [78–81]. An effective coating should meet the requirements of electrochemical compatibility with the zinc electrode and electrolytes and excellent mechanical strength to suppress the growth of zinc dendrites.

Carbon and polymer-based coatings are always chosen for zinc anode protection [82]. For example, the Zn electrodes covered by carbon nanotubes (CNTs) were built [83], where the porous framework can mechanically adjust the volume change and offer a stable electric field during Zn desorption/deposition, hindering the formation and growth of Zinc dendrites and the occurrence of side reactions. With the protection of carbon-based coating, the Zn anode with a stable scaffold exhibited high chemical stability and excellent reversibility, leading to the enhanced ultralong stable cycling lifespan of ZIHC (over 1800 h). Besides carbon-based coating, polymer-based coating with an elastic layer is beneficial to alleviate shape mutation and induce the lateral deposition

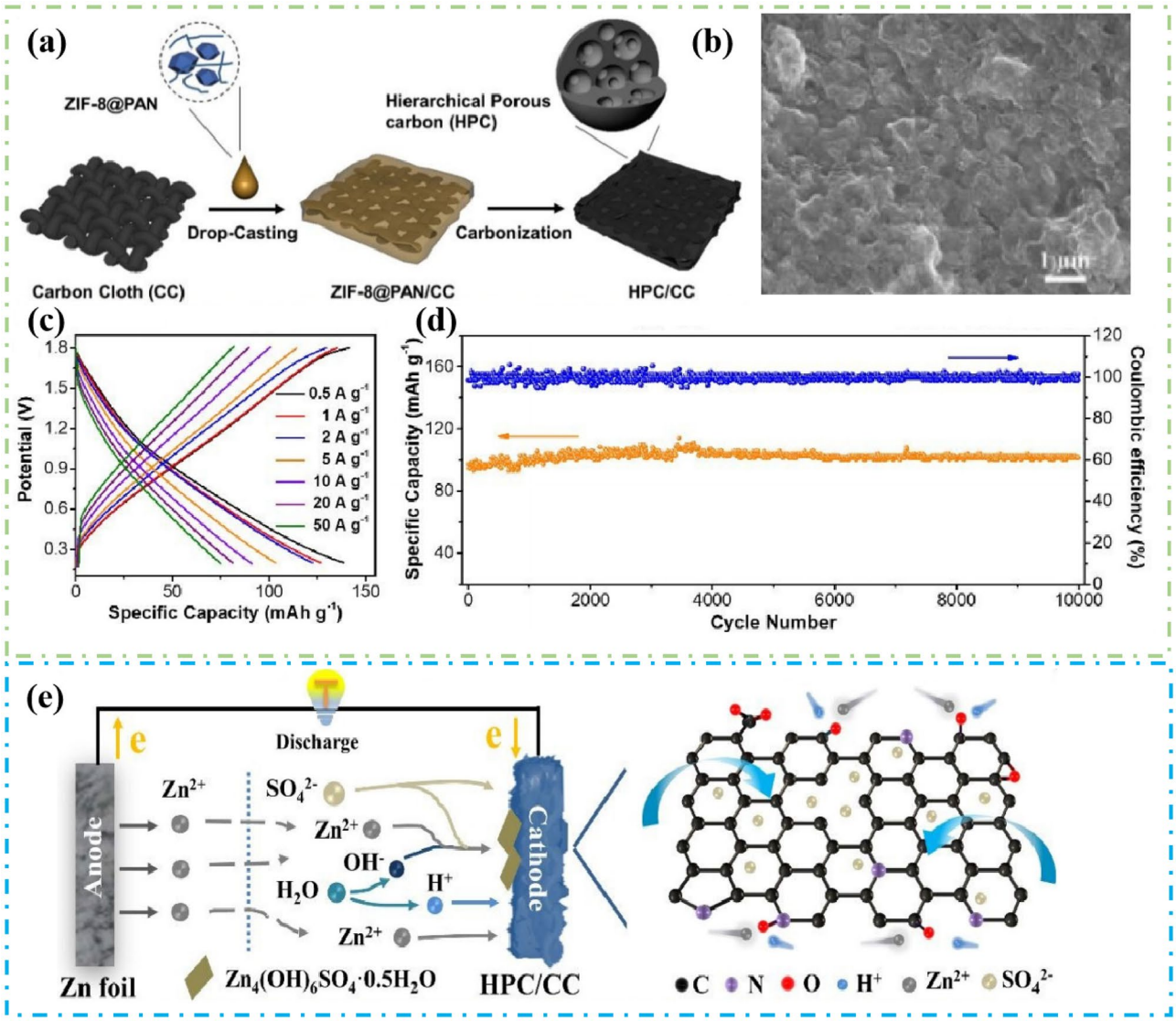

Fig. 9 Morphology and electrochemical properties of HPC/CC: **a** diagrammatic sketch of HPC/CC synthesis; **b** SEM image of HPC/CC; **c** GCD curves of HPC/CC-based ZIHC at 0.5–50.0 A g^{-1}; **d** cycling stability of HPC/CC-based ZIHC at 5 A g^{-1}; (**e**) schematic diagram of ZIHC assembled with HPC/CC. Reproduced with permission from [73], Copyright 2019, RSC

of zinc dendrites [84]. The polyamide films were covered on zinc electrode surfaces by Zhao et al. [85], where the side reactions of zinc ions when passing through the polyamide membrane can be effectively suppressed, thanks to the desolvation of hydrated zinc ions by the polyamide membrane, resulting in satisfactory electrochemical performance. Furthermore, the unique prepared coating limits the diffusion of Zn^{2+} and tunes the deposition process of Zn electrodes due to the coordination of amide groups with Zn^{2+}.

The dominant factor that should be considered in material selection is the affinity with the zinc electrodes when adopting a coating protection strategy. The coating materials with a good affinity for zinc electrodes could effectively tune the electric field, reduce overpotentials, and enhance the uniform deposition on the surface of Zinc, resulting in the enhanced cycling performance of ZIHCs.

4.2 Electrolyte regulation

Electrolyte additives can endow Zn^{2+} diffuse and deposit more uniformly and inhibit side reactions, hindering the formation and growth of zinc dendrites, which have attracted enormous attention in improving the performance of ZIHCs. For example, a superior electrochemical performance ZIHC with multivalent metal ion electrolyte by adding 0.1 M $MgSO_4$ in 2.0 M $ZnSO_4$ electrolyte was reported (Fig. 11a) [86]. The results that proved the multi-advantages of the introduction of Mg^{2+} into the electrolyte were obtained. First, the formation and growth

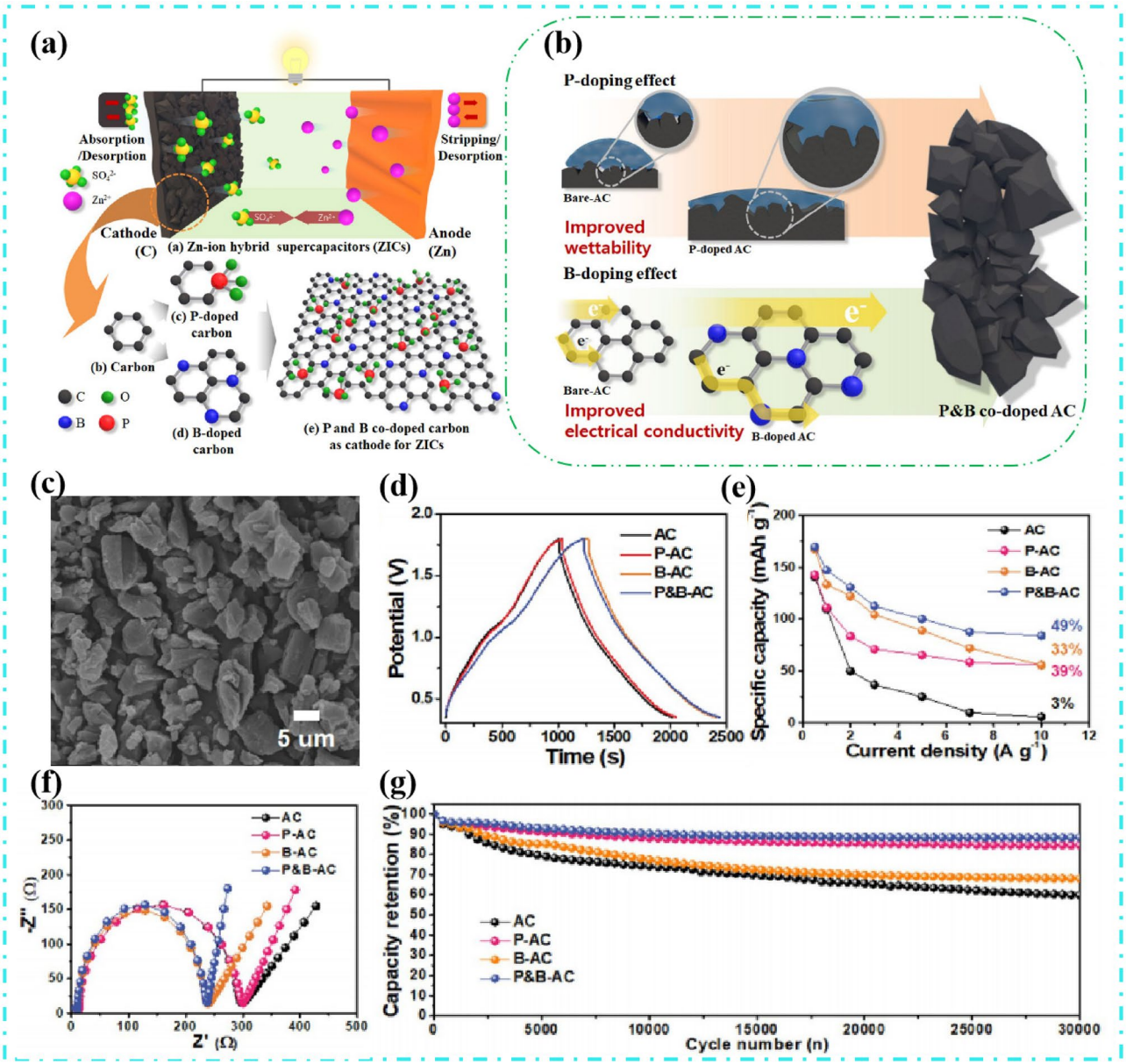

Fig. 10 Morphology and electrochemical properties of P&B-AC and other comparative samples: **a** schematic diagram of ZIHC with P&B-AC as a positive electrode; **b** schematic diagram of the advantages of the optimized P&B-AC performance; **c** SEM image of P&B-AC; **d** GCD curves at 0.5A g^{-1}; **e** specific capacity under different current densities; **f** Nyquist plots; **g** longevity performance for up to 30,000 cycles at 10.0 A g^{-1}. Reproduced with permission from [55], Copyright 2020, ACS

of zinc dendrites on the Zn electrode surface are effectively inhibited, and the side reactions of ion exchange between the Zn anode and electrolyte are weakened (Fig. 11b–e). The ZIHCs with Zn-Mg mixed electrolyte exhibited electrochemical behaviors, including a high capacitance of 154 mAh g^{-1} at 1.0 A g^{-1} (Fig. 11f) and high capacity retention over 98.7% after 10,000 cycles at 5.0 A g^{-1} (Fig. 11g). Mai et al. [87] proposed a high-performance ZIHC (ACC//Zn ZIHC) consisting of activated carbon coat as cathode, metallic Zn as anode, and $ZnSO_4/Na_2SO_4$ solution as electrolyte. The experiment results illustrated that the Zn anode was protected, and the capacitive storage of the ACC cathode was improved due to the intermediate $Zn_4SO_4(OH)_6{\cdot}0.5H_2O$ that was suppressed by adding 1.0 M Na_2SO_4 into 1.0 M $ZnSO_4$ electrolyte. Besides, the voltage window was broadened to 1.9 V, and the energy densities of 100 Wh kg^{-1} and 20 Wh kg^{-1} at power densities of 50.42 W kg^{-1} and 1692 W kg^{-1}, respectively, were achieved in this ZIHCs system.

In addition to cationic electrolyte additives, anionic electrolyte additives have also been identified to effectively improve

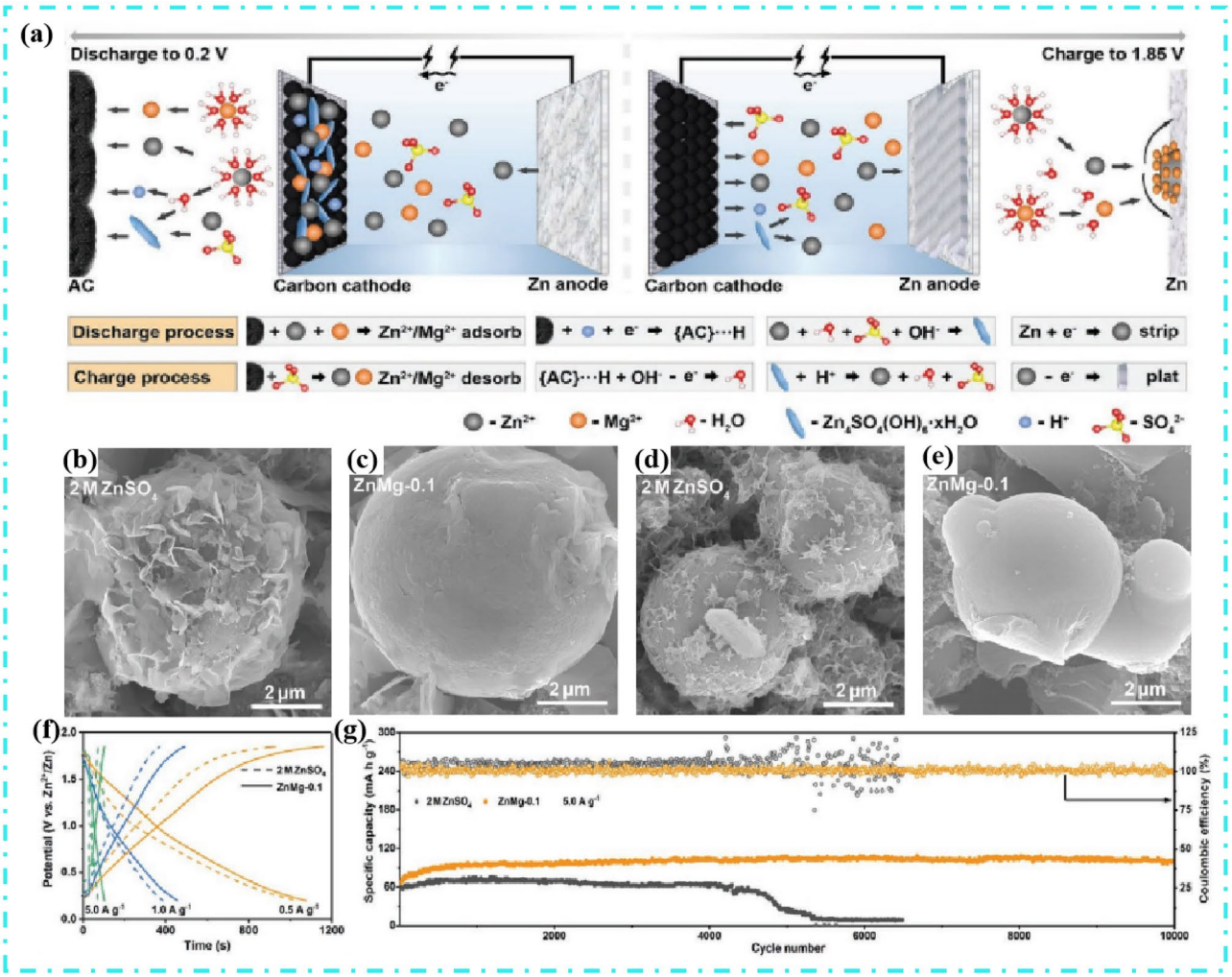

Fig. 11 Morphology and electrochemical properties of AC: **a** schematic diagram of the structure of AC//ZnMg-0.1//Zn equipment and its mechanism of action; **b** SEM image of AC in $ZnSO_4$ electrolyte (discharge state); **c** SEM image of AC in ZnMg-0.1 electrolyte (discharge state); **d** SEM image of AC in $ZnSO_4$ electrolyte (charge state); **e** SEM image of AC in ZnMg-0.1 electrolyte (charge state); **f** GCD curves in $ZnSO_4$ and ZnMg-0.1 electrolyte at 0.5–5.0 A g^{-1}; **g** longevity performance in $ZnSO_4$ and ZnMg-0.1 electrolyte at 5.0 A g^{-1}. Reproduced with permission from [86], Copyright 2021, Wiley–VCH

the electrochemical capacitance of ZIHCs. The ZIHC composed of $ZnSO_4$, $ZnCl_2$, and $ZnAc_2$ as the electrolyte, TiN as cathode, and metallic Zn as anodes was constructed [45] to investigate the consequence of the electrochemical performance of anion carrier. The TiN-SO_4 has the smoothest adsorption process, and the lowest adsorption energy is possessed by SO_4^{2-}, which was proven by discrete Fourier transform (DFT) calculation of the adsorption energies of the TiN cathode for the three anions. Meanwhile, the ZIHC with $ZnSO_4$ electrolyte exhibited the superior electrochemical performance of ultra-high capacitance of 314.5 F g^{-1} at 0.2 A g^{-1} after 10,000 cycles and high capacity retention over 83.92% after resting for 500 h; outstanding stability of the TiN-SO_4 structure improved the self-discharge resistance of the capacitor, which illustrated that capacitance and self-discharge resistance of ZIHCs are significantly affected by different anion types. Currently, electrolyte additives have been effectively applied in improving the electrochemical performance of ZIHCs. Table 2 summarizes the electrochemical properties of electrolyte control in ZIHCs, showing the high efficiency of electrolyte additives in inhibiting side reactions and hindering the formation and growth of zinc dendrites. The unique design of the electrode structure, as well as the modification of the electrolyte, can significantly improve its electrochemical performance. Meanwhile, the choice of suitable electrolytes still leaves room for further exploration.

Table 2 Summary of electrolyte-controlled electrochemical properties in ZIHCs

Materials	Electrolyte	Voltage range (V)	Capacity	Current density (A g^{-1})	Electrochemical performance	Energy density (W h kg^{-1})	Power density (kW kg^{-1})	Ref.
FL-P	0.2 M $ZnCl_2$ in Et_4NBF_4/PC electrolyte	0–2.5	363.9 F g^{-1}	0.2	/	315.6	23.6	[48]
FCNSs	0.2 M $Zn(CF_3SO_3)_2$ in $EMIMCF_3SO_3$	0.1–2.4	300 F g^{-1}	0.1	82% after 35,000 cycles	217	13	[88]
AC	SA-Zn double network hydrogel electrolyte	0–2.4	260.5 mA h g^{-1}	0.2	95.4% after 10,000 cycles	286.6	/	[89]
AC	Redox SA-Zn-Br double network hydrogel	0–2.6	654.8 mA h g^{-1}	2	87.7% after 5000 cycles	605	/	[90]

5 Summary and perspective

In recent years, ZIHCs have been regarded as emerging electronic energy storage devices with good potential and rapid development momentum, combining the powerful cycling stability of traditional supercapacitors while inheriting the superiority of zinc-ion batteries, including abundant resources, environmental protection, high theoretical capacity, and cost-effectiveness. In this work, we report the recent regulated preparation and modification of carbon cathodes for ZIHC (e.g., AC, BC, PC, and HDC); property improvement measures, such as coating, electrolyte ion regulation, electrode structure design to inhibit the formation and growth of zinc dendrites and possible side reactions, have been carried out to improve ZIHCs electrochemical performance.

Although significant breakthroughs have been made in exploring ZIHCs with carbon-based electrodes, the instability of the electrochemical performance and side reactions in the electrolyte system hinder the concrete application of ZIHCs as excellent performance energy storage devices. Therefore, rational structural control and morphology control of carbon-based materials are crucial for high-performance ZIHCs. In addition, the investigation of electrolytes for ZIHCs is now at the very beginning stage; the development of high-performance ZIHCs is inseparable from an electrolyte system with high voltage and excellent stability. As a result, it is worthwhile to strengthen the research on the mechanism of the electrolyte and build a stable electrolyte to reduce the effect of the side reaction of zinc anode.

The future exploration of ZIHCs carbon-based electrodes can be deepened from the following directions. The electrochemical performance of ZIHCs is closely related to the porosity of carbon-based materials. Building a porous structure can enhance the ion adsorption–desorption capacity of electrolytes, which plays an important role in enhancing capacitance and facilitating ion transport. Besides, heteroatom doping is believed to effectively tune the electrochemical structure and surface features of carbon; thus, the electrolyte ions have better migration ability on the electrode surface. Familiarity with the effects and working mechanisms of different dopants and the adoption of accurate and repeatable doping strategies are beneficial for obtaining cathode material with excellent performance. Furthermore, carbon-based composites prepared by compounding carbon with various pseudocapacitive materials are a promising new direction for ZIHCs, which have bigger capacitance and larger operating voltage windows than pure carbon alone, but the problems of low rate and low cycle performance cannot be ignored. Therefore, improving the electrochemical stability of carbon-based pseudocapacitive composites and finding ways to suppress side reactions are still challenges.

Author contribution As per the journal requirement of more than ten contributors, the required contribution can be briefly stated as follows. Yongpeng Ma: conceptualization, methodology, validation, formal analysis, writing – original draft; Chuanxin Hou: conceptualization, writing – original draft, preparation, methodology, writing – review and editing; Hideo Kimura: conceptualization, resources, and writing – review and editing; Xiubo Xie, Huiyu Jiang, Yuping Zhang, and Xueqin Sun: writing – review and editing; Xiaoyang Yang: methodology, validation; Wei Du: supervision, project administration, writing – review and editing.

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Declarations

Conflict of interest The authors declare no competing interests.

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