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# The controlled preparation and performance improvement of mesocarbon microbeads for energy storage

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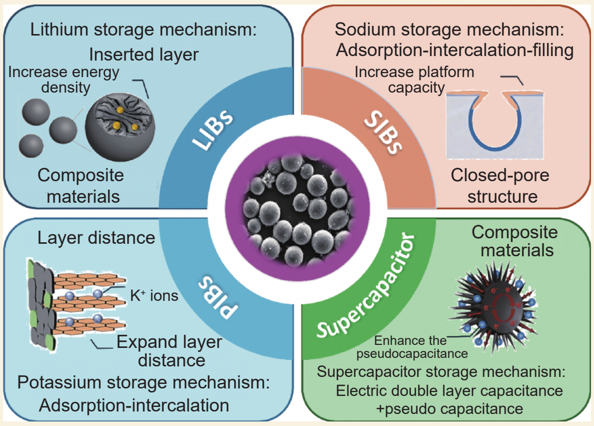
**Abstract:** Mesocarbon microbeads (MCMBs) are a high-performance carbon material that has been widely used in energy storage and as high-temperature structural materials due to their highly controllable microstructure and excellent electrical conductivity. However, with different energy storage mechanisms such as lithium-ion batteries, sodium-ion batteries, potassium-ion batteries, and supercapacitors, MCMBs with a single structure cannot fully meet the different material performance requirements. We review the basic characteristics, preparation methods, formation mechanism and modification strategies of MCMBs, focusing on the relationship between its microstructure and electrochemical performance in various energy storage systems, and its application in other fields. The opportunities and challenges of using MCMBs in different energy storage applications are considered.



**Key words:** Mesocarbon microbeads；Lithium-ion battery；Sodium-ion battery；Potassium-ion battery；Supercapacitor

## 1　Introduction

Mesocarbon microbeads (MCMBs) are an important class of pitch-derived carbon materials formed from polycyclic aromatic hydrocarbons (PAHs)-rich precursors, such as petroleum residues and coal tar pitch. During heat treatment at intermediate temperatures, these precursors undergo thermal cracking, dehydrogenation, and polycondensation under an inert atmosphere, leading to the formation of discotic nematic mesophase domains. Driven by surface tension, these anisotropic domains gradually evolve into spherical entities that can be separated from the isotropic matrix, which are commonly referred to as MCMBs. MCMBs are also described as carbonaceous mesophases or carbonaceous liquid crystals[1−2]. The study of MCMBs originated in the early 1960s, when Brooks and Taylor[3] observed the formation and growth of optically anisotropic microspheres during coal coking. However, their physicochemical nature remained unclear until the early 1970s, when Yamada[4] successfully isolated these spheres from liquid-phase carbonized pitch and formally defined them as MCMBs. Since then, MCMBs have attracted sustained research interest, leading to systematic investigations into their formation mechanism, structural evolution, and functional properties[5]. Owing to their turbostratic layered structure, uniform particle size, and favorable spherical morphology, MCMBs exhibit excellent electrical and thermal conductivity[6], high thermal stability, good self-sintering ability, and mechanical robustness[7−8]. These intrinsic

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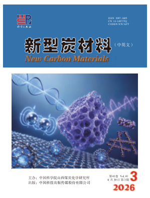

characteristics make MCMBs particularly suitable for electrochemical energy storage. To date, MCMBs have been extensively explored as anode materials for lithium-ion batteries (LIBs)[9–20], and have demonstrated potential in sodium-ion batteries (SIBs)[21–23], potassium-ion batteries (PIBs)[47–48] and supercapacitors[24–29], as well as in high-temperature structural and functional materials, such as carbon/carbon composites and special carbon-graphite materials[30–46].

From a materials design perspective, the electrochemical performance of MCMBs is closely governed by their microstructural characteristics, including sphericity, particle size distribution, carbon layer orientation, interlayer spacing, defect density, and surface chemistry. These parameters can be effectively tailored through precursor selection, additive regulation, and optimization of synthesis strategies, such as thermal polymerization, emulsification and suspension. Rational microstructure engineering enables the simultaneous optimization of electronic conductivity, ion transport kinetics, structural stability, and interfacial compatibility, thereby promoting the practical applications of MCMB-based electrodes in advanced energy storage systems. Accordingly, this review systematically summarizes the fundamental characteristics, formation mechanisms, preparation strategies, state-of-the-art preparation, and applications of MCMBs, with particular emphasis on their structure-performance relationships in LIBs, SIBs, PIBs and supercapacitors.

## 2 Formation mechanisms and morphology of MCMBs

The formation and evolution of MCMBs are intrinsically linked to the microstructure development of carbonaceous mesophase during pitch heat treatment. Generally, aliphatic and aromatic components in pitch undergo thermal polymerization and dehydrogenation, generating large polyaromatic macromolecules under an inert atmosphere at intermediate temperatures. With increasing temperature, the concentration of these macromolecules increases, and lamellar stacking is promoted by van der Waals interactions. To minimize interfacial free energy between isotropic and anisotropic phases, the stacked lamellae aggregate into anisotropic mesophase spheres with reduced surface area. These mesophase spheres subsequently absorb surrounding low-molecular-weight species from the parent matrix and grow through collision, coalescence, accompanied by continued polycondensation, eventually evolving into larger mesophase domains and MCMBs. This mechanism constitutes the classical model for nucleation and growth of MCMBs[49–50]. Despite its conceptual clarity, the classical model is mainly applicable to homogeneous nucleation systems and cannot adequately describe heterogeneous pitch systems containing quinoline-insoluble (QI) components. To address this limitation, Mochida et al.[51] proposed the “microdomain construction” theory, in which mesophase formation proceeds via the initial generation of regularly shaped, sheet-like stacking units. These units subsequently assemble into spherical microdomains, which further evolve into mesophase spheres. This model provides a more reasonable interpretation of growth of mesophase spheres in complex systems. Nevertheless, the idealized assumptions regarding layer size uniformity and stacking order may oversimplify the actual structural complexity of MCMBs. To further clarify the formation mechanism under heterogeneous nucleation conditions, Wang et al.[52] systematically investigated the evolution of MCMBs in coal tar pitch containing a controlled amount of QI components and proposed the “particle basic unit construction” model (Fig. 1a). In this hierarchical assembly mechanism, mesophase formation proceeds through continuous multilevel construction. During heat treatment, the aromaticity of pitch molecules progressively increases, and strong π-π interactions drive the aggregation of aromatic sheets into spherical or granular basic building units. These units subsequently accumulate around mesophase nuclei, promoting the growth of well-defined mesophase spheres and ultimately MCMBs. This model effectively bridges molecular evolution with mesoscale structural development in heterogeneous pitch systems.

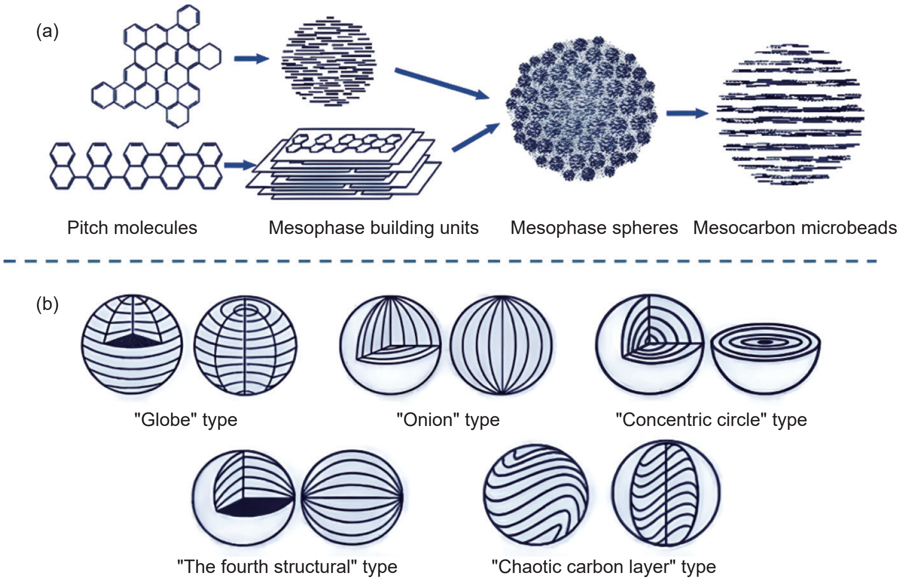

Fig. 1 (a) The “particle basic unit construction” theory for formation of MCMBs. (b) The structural types of MCMBs

The preparation of MCMBs critically depends on precise regulation of the reaction system to control the nucleation density, particle size, and size distribution of mesophase spheres. Typical precursors are heavy PAH-rich feedstocks, including medium-temperature coal tar, ethylene bottom oil, catalytic cracking residues, and their thermally condensed products. The chemical compositions of the precursors including particularly impurity and their content, as well as reaction temperature, system viscosity, and heating history, significantly influence mesophase nucleation, growth, melting, and coalescence behaviors. As a result, MCMBs synthesized under different processing conditions exhibit pronounced diversity in internal microstructure and textural organization. Additionally, in terms of structural characterization, Brooks and Taylor pioneered the systematic investigation of MCMB morphology and proposed the classical Brooks-Taylor structure, also referred to as the “globe” or “latitude” structure. In this model, planar aromatic layers within the sphere exhibit two preferential orientations: parallel and perpendicular to the spherical surface. Subsequent studies have revealed several additional structural configurations of MCMBs (Fig. 1b), including concentric[53], onion-like[54], radial[55] and disordered carbon layer structures[56−57]. In the concentric structure, aromatic carbon layers are uniformly arranged as shells centered on the sphere core. The onion-like structure is characterized by layers predominantly aligned tangentially to the spherical surface, with layer terminations accumulating in polar regions, resulting in short-range order and long-range disorder. In contrast, the radial structure features carbon layers extending outward from the sphere center, with basal planes oriented approximately perpendicular to the surface. MCMBs with disordered carbon layer structures exhibit highly random stacking without a well-defined global orientation, reflecting a low degree of structural ordering.

## 3 Progress in preparation of MCMBs

Since their discovery, MCMBs have been predominantly synthesized through the thermal polycondensation of pitch. With continuous improvements in process control and equipment, this approach has become increasingly mature and remains the most widely adopted route for industrial production. However, the inherent limitations related to particle size distribution, morphological uniformity, and process efficiency have motivated the development of alternative strategies, including the emulsification and suspension methods, as well as several emerging techniques. Representative preparation routes of MCMBs are schematically illustrated in Fig. 2.

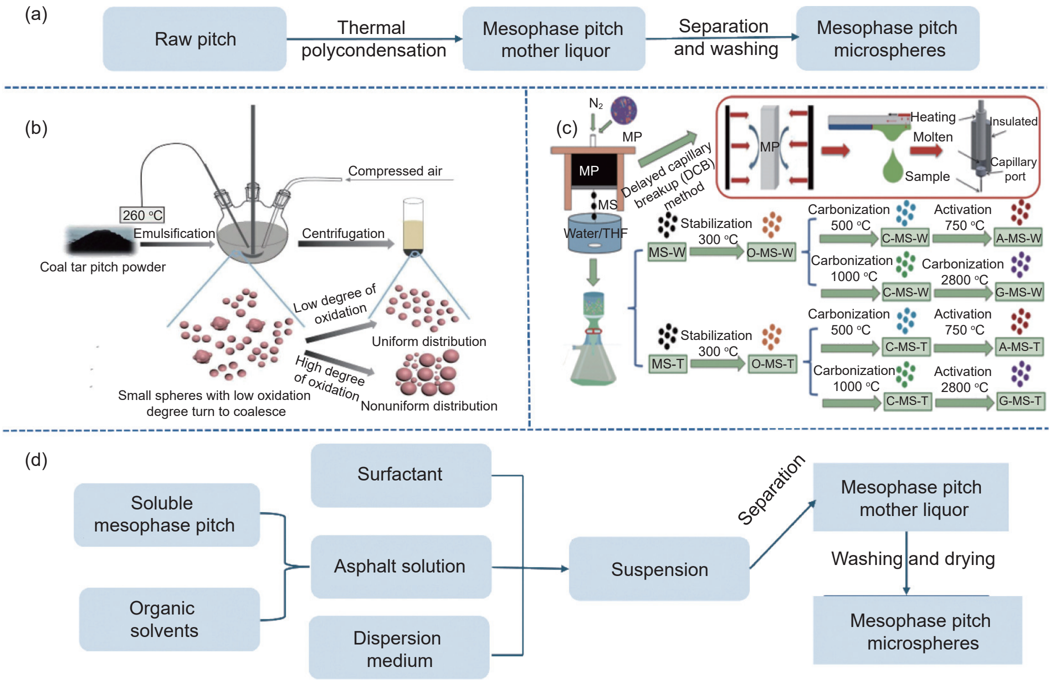

Fig. 2 Schematic diagrams of MCMBs prepared by (a) the thermal polycondensation and (b) the emulsification[58]. Copyright 2020, Elsevier. (c) The delayed capillary breakup[59]. Copyright 2023, Elsevier. and (d) suspension methods

### 3.1 Thermal polycondensation method

The thermal polycondensation method involves the condensation of PAHs into mesophase spheres through heat treatment under an inert atmosphere. These mesophase spheres are dispersed in the pitch matrix and can be isolated to obtain MCMBs. A typical process consists of pitch thermal polycondensation to form mesophase pitch, separation of mesophase microspheres, followed by pre-oxidation and carbonization (Fig. 2a). This method features a relatively simple process, controllable reaction parameters, and good scalability, making it suitable for large-scale production. Nevertheless, uncontrolled nucleation and excessive coalescence often lead to broad particle size distributions and poor morphological uniformity. Numerous studies have demonstrated that precursor composition and molecular structure play decisive roles in mesophase nucleation and growth. Among these factors, the content of QI species is particularly critical. Gu et al.[60] reported that high-quality MCMBs with good sphericity and narrow size distribution could be obtained when the primary QI (PQI) content in high-temperature coal tar pitch was maintained within 7%–13%, yielding up to 33.09%. Within this range, PQI serves as effective heterogeneous nucleation sites. Insufficient PQI limits nucleation density, whereas excessive PQI disrupts molecular orientation and suppresses sphere growth. To regulate QI content, catalytic conversion strategies have been explored. For instance, Zhang et al.[61] employed $AlCl_3$ to catalyze the polycondensation of heavy coal tar, reducing QI content while promoting mesophase formation and achieving an MCMB yield of 17.91%. Alternatively, co-polycondensation with reactive additives has proven effective. Yan et al.[62] introduced coal pyrolysis extract (CPE) into coal tar pitch, where oxygen-containing functional groups served as active sites to facilitate MCMB formation. An optimal CPE addition of 3% improved sphericity and reduced surface defects. Similarly, the addition of 7% ethylene tar pitch increased the MCMB yield to 32.7% while enhancing particle size uniformity[63]. The deliberate introduction of solid additives provides another means of regulating MCMB formation, though their effects vary significantly. Yang et al.[64] found that rosin acted as an effective nucleating agent, increasing the MCMB yield to 54.6% by promoting sphere aggregation while suppressing excessive fusion. In contrast, inert additives such as micron-sized alumina particles disrupted carbon layer orientation and inhibited sphere growth, resulting in reduced yields[65]. Easily graphitizable materials, such as natural flake graphite, can act as nucleation centers but often increase textural complexity[66]. By comparison, organic additives, including biomass-derived tar pitch, exploit their high oxygen content to enhance cross-linking and modulate interlayer spacing and defect structures in MCMBs[67]. Additionally, following thermal polycondensation, MCMBs must be separated from the pitch matrix. Common separation techniques include solvent extraction and centrifugation. Solvent separation typically involves solvent addition, heating, stirring, vacuum filtration, and Soxhlet extraction, with the resulting MCMB morphology being strongly solvent-dependent. More recently, supercritical extraction has emerged as an alternative. Zhang et al.[68] employed supercritical fluid fractionation to selectively remove components unfavorable for sphere formation, thereby improving MCMB quality.

### 3.2 Emulsification method

The emulsification method represents an indirect but more advanced strategy for MCMB preparation. In this approach, mesophase pitch is first obtained by thermal polycondensation and subsequently emulsified in a dispersion medium to generate spherical mesophase domains, which are converted into MCMBs through carbonization. A typical procedure includes dispersing mesophase pitch in a suitable medium, heating and stirring to form an emulsion, cooling to obtain a suspension, followed by centrifugation, washing, drying, and carbonization. For example, Yuan et al.[58] proposed a simultaneous spheroidization and stabilization strategy (Fig. 2b), in which medium-temperature coal tar pitch was emulsified under an air atmosphere. Simultaneous oxidation and cross-linking stabilized the microspheres in a single step, eliminating the need for a separate pre-oxidation step and significantly simplifying the process. The choice of dispersion medium plays a decisive role in this method. Li et al.[69] compared glycerol and high-temperature-resistant silicone oil as dispersion media and found that glycerol promoted microsphere adhesion, whereas silicone oil, owing to its chemical inertness and thermal stability, effectively stabilized dispersion. Consequently, silicone oil was adopted in both spheroidization and stabilization processes, where controlled air introduction facilitated cross-linking. Compared with the thermal polycondensation method, the emulsification method generally produces MCMBs with higher degrees of polycondensation, narrower particle size distributions, lower light-component content, and fewer impurities, leading to improved performance. Moreover, the simultaneous stabilization in emulsification expands precursor types to low-softening-point pitches and enhances carbonization yield. However, conventional emulsification processes often involve complex operations, non-melting treatments, and stringent process control. To address these issues, Korai et al.[70] proposed a method using medium-temperature pitch itself as the emulsifying medium, thereby eliminating external media such as silicone oil and simplifying post-treatment steps.

### 3.3 Suspension method

In the suspension method, mesophase pitch is dissolved in an organic solvent and dispersed into water or another immiscible medium with the aid of surfactants. Under controlled stirring and heating, mesophase pitch microspheres are formed, which are subsequently recovered by filtration and converted into MCMBs through pre-oxidation and carbonization. The process flow is schematically shown in Fig. 2d. Yoon et al.[71] demonstrated that suspension medium properties, surfactant type and concentration, pitch solution concentration, and stirring conditions critically influence microsphere size and morphology. Compared with emulsification, the suspension method effectively suppresses microsphere agglomeration through surfactant stabilization and enables finer control over particle size. However, its applicability is limited by the requirement for soluble mesophase pitch and the complexity of parameter optimization. In addition, the resulting microspheres are typically non-melted, indicating insufficient internal structural rearrangement.

### 3.4 Other emerging methods

To overcome the limitations of traditional

MCMBs preparation routes in terms of process complexity, cost control, and particle size regulation, researchers have continuously explored new preparation strategies. Among them, Dong et al.[59] innovatively proposed the delayed capillary breakup (DCB) method for preparation of MCMBs based on the dripping behavior and rheological properties of pitch melt spinning process (Fig. 2c). In this method, the mesophase pitch containing mesophase spheres is melted and extruded into the receiving medium through a capillary nozzle to directly form pitch microspheres, and then pre-oxidized and carbonized to obtain MCMBs. On this basis, Zhang et al.[72] further used tetrahydrofuran (THF) to control the extraction of mesophase pitch microspheres prepared by the DCB method to regulate their molecular structure. The DCB method has achieved an important breakthrough in concept and provides a promising technical path for the controllable and continuous preparation of MCMBs.

Overall, MCMB preparation strategies can be classified into thermal polycondensation, emulsification, suspension, and emerging processes, each exhibiting distinct advantages and limitations as summarized in Table 1. Thermal polycondensation is the most mature and scalable approach but requires strict control over nucleation and coalescence to achieve uniform particles. Emulsification enables precise regulation of microsphere morphology and size distribution, yielding MCMBs with higher structural order, albeit with increased process complexity. The suspension method suppresses agglomeration and allows fine particle size control but is constrained by precursor solubility and incomplete sphere melting. Emerging strategies, such as the DCB method, offer promising routes toward process simplification and continuous manufacturing. Ultimately, rational selection and integration of preparation methods, guided by precursor chemistry and target microstructural requirements, are essential for tailoring MCMBs for advanced energy storage and other applications.

**Table 1 Comparison of preparation methods for MCMBs**

| Feature | Method | | | |
|---|---|---|---|---|
| | Thermal polycondensation | Emulsification | Suspension | Emerging methods (e.g. DCB) |
| Advantage | Simple, controllable parameters | Narrow PSD, high purity, enables synchronous processing | Excellent size control, suppresses agglomeration | Simplified process, continuous production potential |
| Limitation | Broad particle size distribution, poor uniformity, post-separation required | Complex operation, stringent control, external media often required | Requires soluble pitch, complex optimization | Mostly developmental, maturity and applicability need validation |
| Scalability | High | Medium | Medium to low | Promising |
| Yield/% | 33[60], 18[61], 33[63], 36–55[64], 12–15[65] | 65–75[58], 86–92[69], 20–30[70] | / | 74[58], 76[72] |
| Particle size/μm | 16[60], 24[62], 20[63], 12–24[64], 4–15[65] | 0.2–4.5[58], 20–40[69], 1–10[70] | 2[71] | 1–2[59, 72] |
| Graphitization degree | 0.777[60], 0.842[61], 0.834[62], 0.799[63] | $d_{002}$=0.372 nm[58] (1000 °C) | / | 0.895[59], 0.902[72] |

## 4 Applications of MCMBs

Benefiting from their controllable carbon-layer structure, spherical morphology, and high physicochemical stability, MCMBs have been widely applied in energy storage and advanced carbon-based systems (Fig. 3). Beyond serving as precursors for high-density carbon/graphite materials and catalyst supports[73–75], MCMBs have emerged as versatile anode materials for LIBs, SIBs, PIBs and supercapacitors. Their favorable electrical conductivity, high tap density, structural robustness, and tunable microstructure enable them to serve not only as active materials but also as conductive frameworks and structural scaffold.

### 4.1 Anode materials for LIBs

As a representative spherical carbon material with a relatively ordered carbon-layer structure, MCMBs have long been regarded as promising anode candidates for LIBs. Early studies primarily focused on elucidating the relationship between intrinsic carbon texture and electrochemical behavior. For example, Chang et al.[55] systematically compared MCMBs with different carbon-layer arrangements and demonstrated that the samples featuring longitude-like

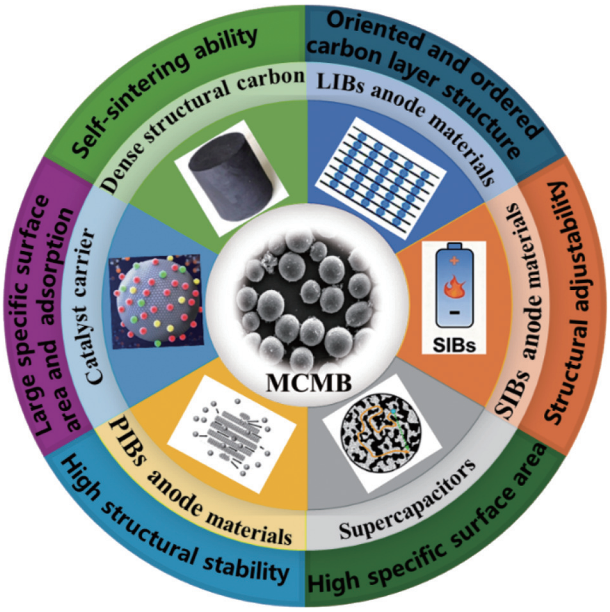

Fig. 3 The applications of MCMBs as a multifunctional anode material platform and advanced carbon-based systems

textures with dislocation defects exhibited superior cycling stability, delivering discharge capacities of 282–300 mAh $g^{-1}$. This performance enhancement can be attributed to the unique carbon-layer orientation. The longitude-like texture provides more accessible edge sites and shortens the solid-state diffusion pathways for $Li^+$ insertion/extraction, while the dislocation defects introduce additional active sites and buffer the volume changes during cycling, thereby improving both the reversible capacity and structural stability. These findings established a fundamental basis for the structural design of anodes based on MCMBs. Subsequent research has emphasized surface engineering as an effective strategy to further enhance the electrochemical performance. Lin et al.[76] proposed a simple solution suspension coating method, and successfully prepared glucose-derived carbon-coated MCMBs (G-MCMBs) by constructing a uniform amorphous carbon layer with controllable thickness on the surface of MCMBs. As shown in Fig. 4a, a thin (~4 nm) coating effectively passivated surface defects while introducing ion-transport-favorable micropores, leading to a significant increase in reversible capacity from 295 to 347 mAh $g^{-1}$ and excellent cycling stability over 150 cycles. This work highlighted the effectiveness of nanoscale surface modification in simultaneously improving capacity and kinetic performance.

To meet the increasing demand for high-energy-density LIBs, MCMBs have been employed as conductive and mechanically robust frameworks for high-capacity active materials, particularly silicon. Fan et al.[77] constructed MCMBs/nano-Si/carbon sandwich structures, in which MCMBs acted as conductive skeletons to buffer silicon volume expansion, achieving a

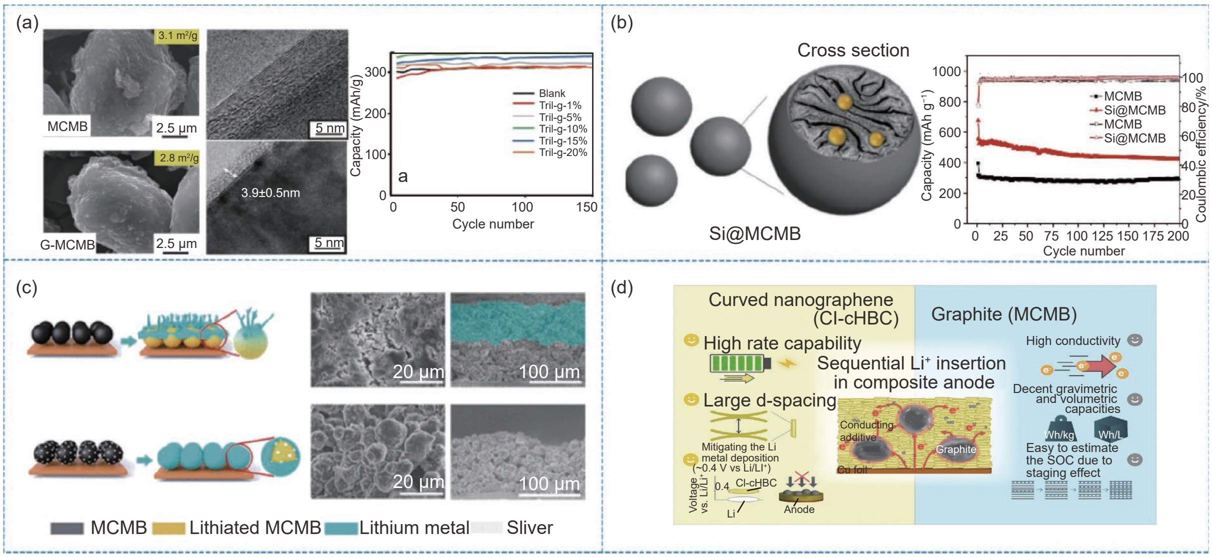

Fig. 4 (a) SEM, TEM images and cycle performance curves of pristine MCMBs and G-MCMBs[76]. Copyright 2022, Elsevier. (b) Composite structure design of Si and MCMBs and its cycle performance curve[78]. Copyright 2022, Elsevier. (c) Schematic diagram of the lithium plating process of MCMBs and @Ag-75, as well as SEM images of the surface and cross-section after 30 cycles[79]. Copyright 2022, American Chemical Society. (d) A schematic diagram of the synergistic effect of Cl-cHBC and MCMBs in the composite anode[80]. Copyright 2025, John Wiley and Sons capacity retention of 90.3% after 100 cycles. Building on this concept, Du et al.[78] further embedded silicon nanoparticles into the disordered carbon layer of MCMBs to construct Si@MCMBs composites. In Fig. 4b, the internal carbon layers accommodated silicon expansion while maintaining electrical connectivity, enabling a reversible capacity of 421.1 mAh $g^{-1}$ after 200 cycles at 0.2 A $g^{-1}$. More recently, MCMBs have been extended to interfacial regulation in lithium-metal-related systems and the construction of hybrid anodes. Dong et al.[79] introduced silver nanoparticles onto MCMBs surfaces to create lithiophilic sites that guide uniform lithium nucleation and deposition (Fig. 4c), effectively suppressing dendrite growth and improving cycling stability at low N/P (anode lithiation capacity/cathode delithiation capacity) ratios. In addition, Cha et al.[80] combined MCMBs with Cl-substituted contorted hexabenzocoronene (Cl-cHBC) to construct a hybrid anode with a “sequential insertion” mechanism. As shown in Fig. 4d, the complementary lithium storage potentials and transport kinetics of the 2 components enabled high-rate capability and long-term cycling stability. Overall, MCMBs have evolved from model graphite anodes into multifunctional LIB anode platforms through intrinsic structural optimization, surface modification, and rational composite design.

### 4.2 Anode materials for SIBs

As a typical soft carbon material, MCMBs have been systematically explored for SIBs, with research progressing from fundamental sodium-storage mechanisms to rational multiscale structural design[81]. Early studies demonstrated that sodium storage in MCMBs is dominated by reversible $Na^+$ intercalation into graphite-like microcrystalline layers (Fig. 5a). MCMBs carbonized at 700 °C exhibited an expanded interlayer spacing of 0.357 nm, enabling a reversible capacity of 208 mAh $g^{-1}$, albeit with moderate initial coulombic efficiency (ICE) due to irreversible sodium loss and sluggish diffusion kinetics[82]. In contrast, MCMBs carbonized at 800 °C, where stacked graphene microcrystallites with an interlayer spacing of 0.347 nm facilitated more reversible sodium insertion/extraction. These samples delivered enhanced cycling stability, maintaining capacities of ~ 180 mAh $g^{-1}$ after 100 cycles with coulombic efficiencies approaching 99%[83].

To further improve sodium-storage performance, extensive efforts have been devoted to structural regulation through chemical and physical modification. Surface treatments such as NaOH etching[84] and air oxidation[85] introduce defects, mesopores, and expanded interlayer spacing, thereby enhancing ion transport and activating additional storage sites. As shown in Fig. 5b, NaOH etching markedly increased the surface roughness of MCMBs and generated abundant ion-transport channels, thereby significantly improving sodium-storage kinetics. As a result, the etched MCMBs exhibited substantially enhanced initial discharge/charge capacities of 441/249 mAh $g^{-1}$ at 15 mA $g^{-1}$. This chemical etching process not only introduces surface defects and mesoporous features but also partially expands the interlayer spacing of carbon microcrystallites, enabling a synergistic sodium-storage mechanism that combines bulk-phase $Na^+$ intercalation with surface-controlled capacitive contributions. Similarly, air oxidation treatment effectively modified the carbon framework through the introduction of oxygen-containing functional groups, which promoted cross-linking of the carbon skeleton while suppressing excessive structural ordering during subsequent heat treatment. This dual effect leads to further expansion of interlayer spacing and an increased defect density, thereby facilitating $Na^+$ diffusion and activation of additional storage sites. Consequently, the oxidized MCMBs delivered a reversible capacity of 286 mAh $g^{-1}$ at 25 mA $g^{-1}$. In addition to post-synthesis surface modification, co-carbonization with biomass-derived precursors provided an alternative route to structural optimization. The resulting MCMBs exhibited outstanding cycling durability, with a capacity retention rate of 99.1% after 70 cycles, demonstrating the effectiveness of crosslinked carbon frameworks in stabilizing sodium storage behavior[67].

Recent advances emphasize cross-scale synergistic regulation of particle size and microcrystalline

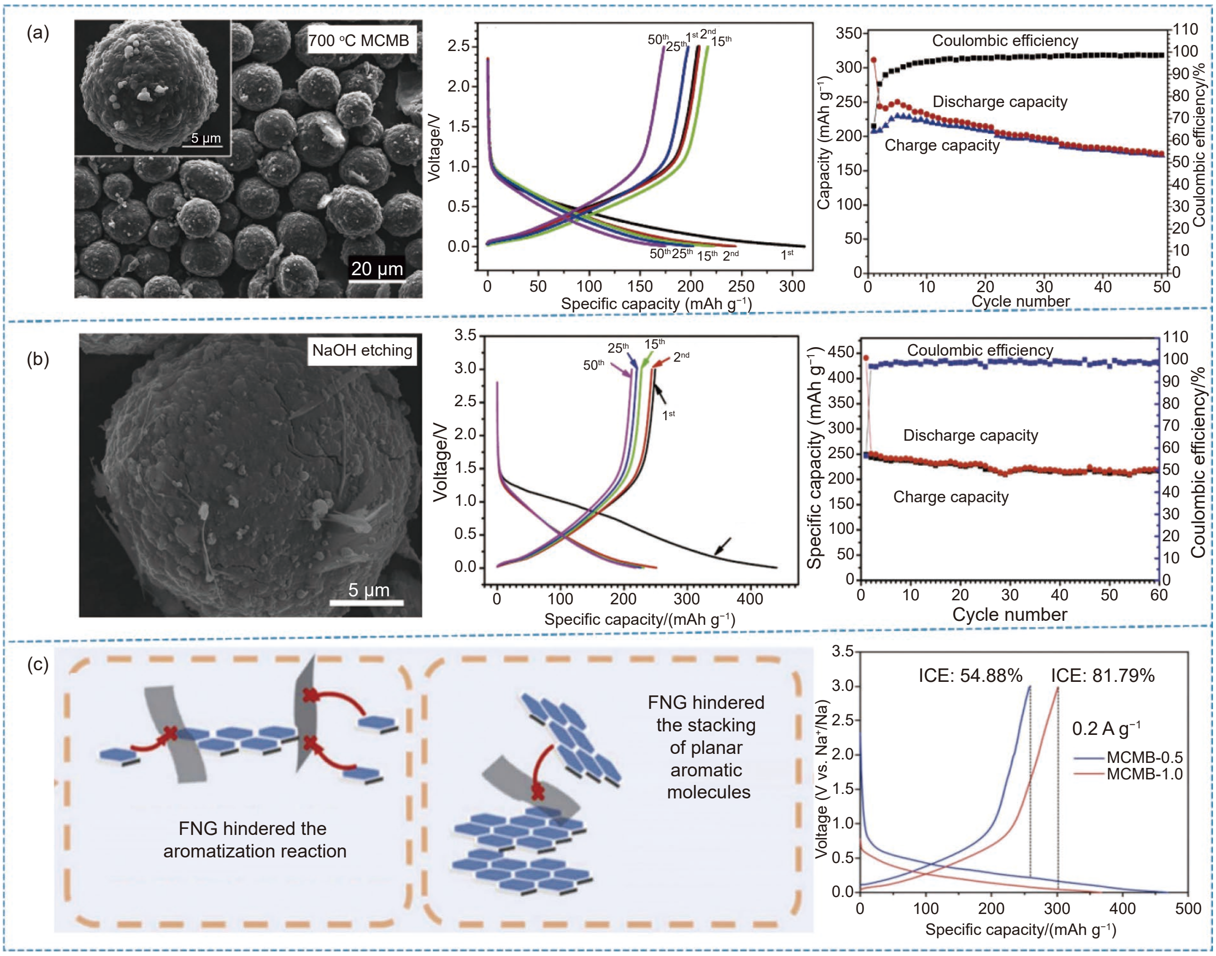

Fig. 5 (a) SEM images, charge-discharge curves and cycle performance curves of MCMBs carbonized at 700 °C[82]. Copyright 2015, Elsevier. (b) SEM image, charge-discharge curve and cycle performance curve of MCMBs etched by NaOH[84]. Copyright 2021, Elsevier. (c) Schematic diagram of the effect of FNG on the structure of MCMB crystallites and the corresponding charge-discharge curves[86]. Copyright 2023, Elsevier

structure. The introduction of functionalized graphene (FNG) during synthesis enables the formation of ultra-large MCMBs (~250 μm) with reduced surface area and expanded interlayer spacing (~0.3566 nm) (Fig. 5c)[86]. FNG has a dual role in the synthesis. On the one hand, it acts as a physical barrier to prevent the fusion of mesophase spheres, thereby successfully preparing an ultra-large MCMBs with a particle size of about 250 μm, reducing the active specific surface area of the electrode and helping to form a stable solid electrolyte interface film. On the other hand, the surface functional groups interfere with the orderly stacking of aromatic molecules, which promotes the increase of interlayer spacing of 0.3566 nm and the formation of long-range disordered microcrystalline structure. This "size-lattice" collaborative design improves the reversible capacity to 300 mAh g$^{-1}$, interface stability, Na$^+$ diffusion kinetics, and the ICE to 81.79%, making a transition toward multi-parameter optimization of MCMB-based SIB anodes.

### 4.3 Anode materials for PIBs

Owing to their spherical morphology, layered structure, and multidirectional ion-transport pathways, MCMBs exhibit distinct advantages as anode materials for PIBs[87]. Compared with natural graphite (NG) and artificial carbon-coated graphite (CG), MCMBs show superior long-term cycling stability. As shown in Fig. 6a, the initial specific capacity of MCMBs was 197.0 mAh g$^{-1}$ at a current density of 0.1 A g$^{-1}$, still maintaining 173.0 mAh g$^{-1}$ after 500 cycles[88]. In addition, potassium-storage behavior in MCMBs is strongly influenced by electrolyte chemistry. In carbonate-based electrolytes, MCMBs favor stepwise intercalation of bare K$^+$ ions and reversible formation of

KC$_8$, resulting in high capacity and low discharge plateau (~ 0.2 V). In contrast, ether-based electrolytes promote solvated-ion co-intercalation, which induces structural degradation and rapid capacity decay Fig. 6b[89]. The different behaviors arise from the interlayer spacing of MCMBs (~ 0.34–0.36 nm), which is sufficient for the insertion of desolvated K$^+$ but insufficient to accommodate the much larger solvated K$^+$ complexes. The co-intercalation of solvated ions in ether electrolytes exerts severe exfoliation stress on the carbon layers, leading to structural collapse. Surface engineering provided an effective route to enhance PIB performance. Mild KOH activation can generate funnel-shaped, locally expanded interlayer structures on MCMB surfaces (Fig. 6c), accelerating K$^+$ diffusion while preserving bulk graphitic order. The KOH-modified MCMBs (K$_3$C-900) exhibited a higher reversible capacity of 254 mAh g$^{-1}$

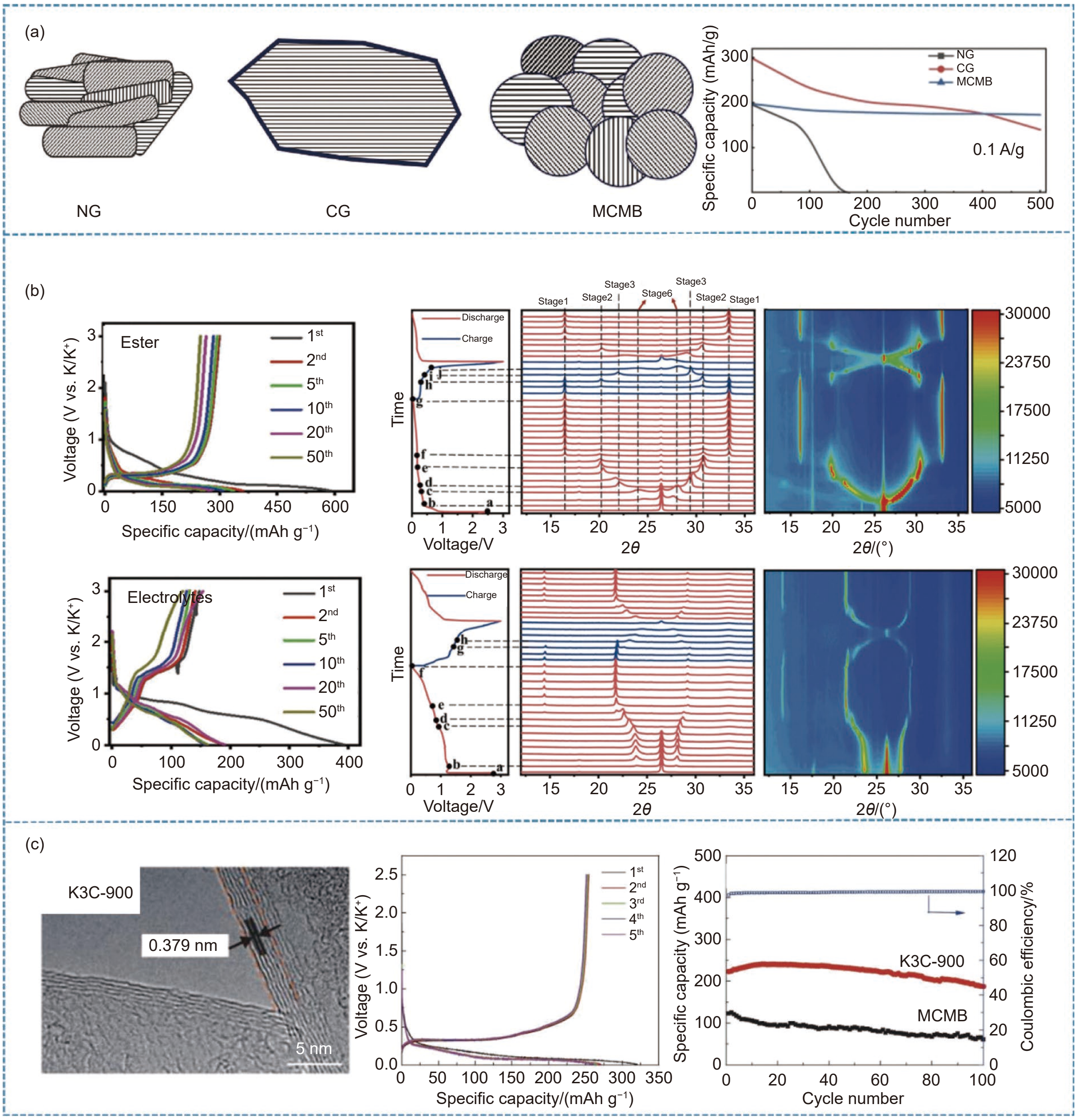

Fig. 6 (a) A schematic diagram of NG, CG, MCMBs samples and their cycle performance curves[88]. Copyright 2024, Springer Nature. (b) The charge-discharge curves and corresponding in-situ XRD patterns of MCMB electrodes in ester (0.8 mol L$^{-1}$ KPF$_6$ in EC/DEC (1 : 1, *v*/*v*)) and ether electrolytes (1.0 mol L$^{-1}$ KTFSI in tetraglyme)[89]. Copyright 2021, John Wiley and Sons. (c) TEM image, charge-discharge curves and cycle performance curves of KOH-modified MCMBs[90]. Copyright 2023, Elsevier at 0.05 A $g^{-1}$, and its capacity retention rate was significantly better than that of the original MCMBs after 100 cycles at 0.1 A $g^{-1}$, demonstrating the importance of interfacial structure regulation in potassium storage[90].

## 4.4 Electrode materials for supercapacitors

MCMBs also show considerable promise as supercapacitor electrode materials due to their spherical geometry, good electrical conductivity, and structural stability. Through oxidation and thermal expansion, pristine MCMBs can be transformed into extended MCMBs with hierarchical porosity and enlarged interlayer spacing (Fig. 7a), which facilitate ion diffusion and intercalation, thereby enhancing capacitance performance[91]. In specific electrolytes, such as spiro quaternary ammonium salts, MCMB-based electrodes exhibit ion-intercalation-dominated energy storage behavior with excellent cycling stability[92]. To further increase capacitance, heteroatom doping and pseudocapacitive material integration have been widely adopted. Sulfur-doped MCMB-derived graphene exhibits improved wettability, conductivity, and reversible redox activity, resulting in enhanced capacitance and durability[93]. Core-shell architectures based on MCMB substrates, such as $NiCo_2S_4$@MCMB composites (Fig. 7b), effectively combine electric double-layer capacitance with faradaic pseudocapacitance, delivering a high specific capacitance as high as 936 F $g^{-1}$ at a current density of 1 A $g^{-1}$ and outstanding cycling stability[94]. Similar synergistic effects were reported for $NiCo_2O_4$@MCMB composites[95].

In addition, multicomponent carbon architectures integrating spherical MCMBs with 10 graphitized nanofibers (GNFs) and 20 mesoporous carbons (MCs), namely MCMB/GNF/MC composite Fig. 7c, enabled synergistic optimization of packing density, conductivity, and accessible surface area, leading to improved electrochemical performance with a specific capacitance of 393 F $g^{-1}$ at a current density of 1 A $g^{-1}$[96]. Compositing MCMBs with conductive polymers is also an effective way to enhance conductivity and introduce pseudocapacitance. For example, a uniform core-shell structure of PANI/ACMB composite was formed by in-situ coating polyaniline (PANI) on the surface of activated MCMBs (ACMBs). As shown in Fig. 7d the PANI/ACMB composite combined the Faraday pseudocapacitance of PANI and the electric double layer capacitance of ACMBs, so that the specific capacitance of PANI/ACMB (433.75 F $g^{-1}$) was much higher than that of ACMBs

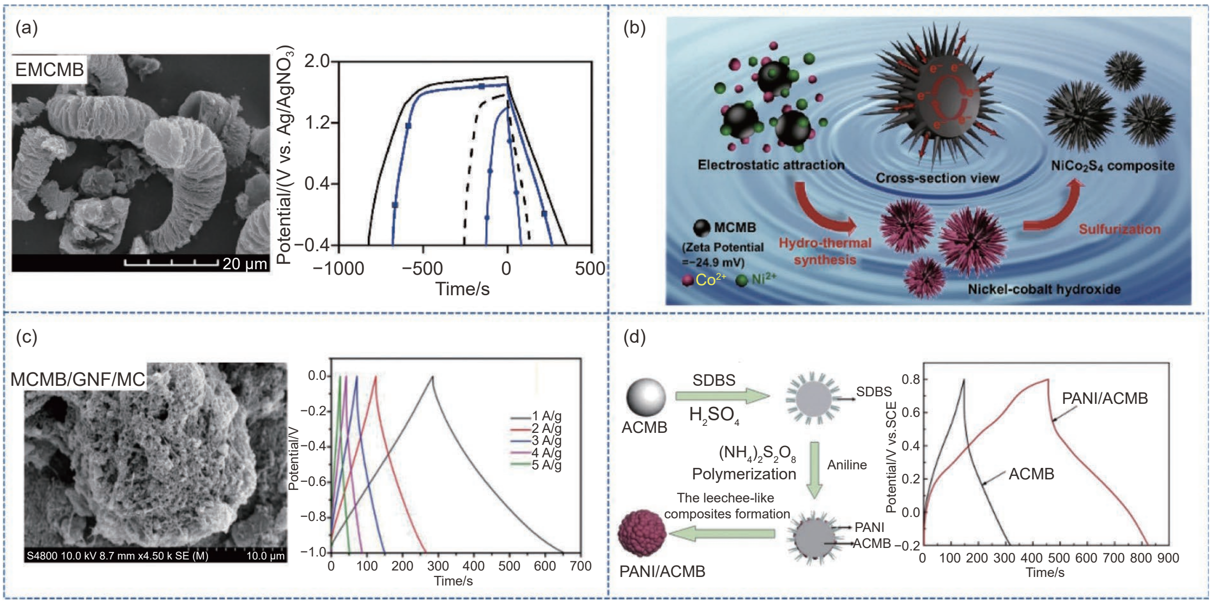

Fig. 7 (a) SEM image and the charge-discharge curves of the extended EMCMBs[91]. Copyright 2020, Elsevier. (b) Schematic diagram of the synthesis process for the urchin-like $NiCo_2S_4$@MCMB composite[94]. Copyright 2019, Springer Nature. (c) SEM image of the MCMB/GNF/MC composite and its corresponding charge-discharge curves[96].Copyright 2025, MDPI. (d) Schematic illustration of the preparation process for PANI/ACMB composites, and the charge-discharge curves of ACMBs and PANI/ACMB composites[97]. Copyright 2012, Elsevier

(156.25 F $g^{-1}$)[97]. The porous structure of ACMBs not only contributes to electric double layer capacity but also provides a large surface area and anchoring sites for the uniform deposition of the PANI layer. This uniform coating ensures intimate contact between the two components, maximizing the utilization of pseudocapacitance from PANI and facilitating electron transfer from PANI to the conductive MCMB core.

### 4.5 Other applications

Beyond energy storage, MCMBs have been expanded to a range of advanced applications. In structural ceramics, MCMB-reinforced SiC composites exhibited high strength and low friction, suitable for extreme aerospace environments[98]. Incorporation of MCMBs into $Al_2O_3$ matrices has enabled efficient microwave absorption through dielectric regulation[99]. In electrocatalysis, $MnO_2$/MCMB composites enhanced oxygen reduction activity for solid-state zinc-air batteries[100]. Moreover, 3D graphitized MCMB networks have been developed as thermal interface materials with high in-plane thermal conductivity for advanced thermal management applications[50]. Collectively, these studies demonstrated the multifunctional nature of MCMBs enabled by rational structural and composite design.

## 5 Conclusions and prospects

MCMBs have emerged as versatile and competitive carbon materials for energy storage, owing to their spherical morphology, tunable microstructure, high conductivity, and structural stability. In LIBs, MCMBs have evolved from conventional graphite anodes into multifunctional platforms. Through surface engineering and hybridization with high-capacity materials (e.g., Si, Sn), MCMB-based anodes achieve enhanced energy density, rate capability, and cycling stability. In SIBs and PIBs, targeted microstructural modifications, such as expanded interlayer spacing, defect engineering, and porosity control, effectively mitigate sluggish alkali-ion kinetics, highlighting the promise of MCMBs as next-generation anodes. In supercapacitors, MCMBs serve either as electric double-layer materials or as conductive scaffolds for pseudocapacitive components (e.g., transition metal oxides), offering pathways toward electrodes with both high power and high energy density.

Despite substantial progress, fully exploiting the structural advantages of MCMBs requires further efforts. A critical assessment of their inherent trade-offs is essential for rational design. For instance, high graphitization benefits conductivity and long-term cycling in LIBs but reduces defect sites and interlayer spacing, two key factors for capacitive storage and ion insertion in SIBs and PIBs. Balancing these properties remains a fundamental, application-specific challenge. Moreover, the scalability and economic viability of the emerging synthesis methods remain uncertain compared with conventional coal-tar pitch or naphthalene-based processes, necessitating comprehensive life-cycle and cost-benefit analyses. Relative to other carbon anodes, such as low-cost but less conductive hard carbon, high-performance yet expensive graphene and carbon nanotubes, MCMBs occupy a middle ground, offering a unique combination of spherical morphology, moderate conductivity, and relatively mature production technology. Thus, their cost-effectiveness must be evaluated within the context of specific performance requirements for each energy storage system.

Future research directions are summarized in Fig. 8 and outlined below.

(1) LIBs: MCMBs are expected to play a key role as structural substrates in advanced composite anodes. Emphasis should be placed on interface regula-

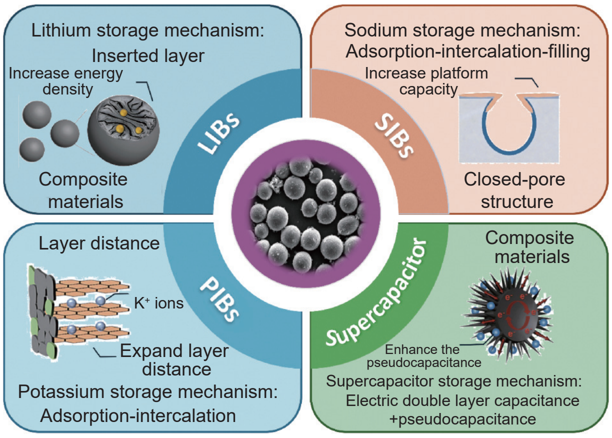

Fig. 8 The design strategy of MCMBs for different energy storage application fields in future

tion and collaborative optimization of ion transport and reaction kinetics to achieve high rate performance and long-term cycle stability. Strategies to reduce volume expansion in high-capacity composites, such as building a strong and flexible carbon shell, will be crucial. A key trade-off here is how to introduce sufficient buffering capacity to accommodate volume changes while maintaining a high degree of graphitization to meet the conductivity requirements.

(2) SIBs: MCMBs are mainly studied as soft carbon precursors. Controlled conversion into hard carbon through cross-linking agent or oxidation strategy, combined with closed-shell engineering, is crucial for improving reversible sodium storage capacity and initial Coulombic efficiency. However, this transformation is usually at the expense of sacrificing conductivity and increasing structural disorder. Future work must focus on precisely controlling pore structures to maximize sodium storage while avoiding serious damage to electron transport or excessive electrolyte decomposition.

(3) PIBs: Targeted microstructural modifications, including interlayer spacing expansion, defect engineering, and stacking sequence optimization, are essential to promote synergistic adsorption/embedded storage mechanisms and improve potassium storage performance. The challenge is to achieve a sufficiently large interlayer spacing to accommodate large potassium ion insertion, while maintaining sufficient structural integrity to prevent spalling and capacity decay during repeated cycling. The stability of these expansion structures under long-term cycling and high voltage is still a key issue and needs to be further studied.

(4) Supercapacitors: MCMBs are mainly used as conductive skeletons and activated carbon precursors. Constructing hierarchical porous structures, heteroatom doping, and compositing them with pseudocapacitive materials are effective strategies to achieve a balance between high capacitance, high power density, and ultra-long cycle life. The main limitation lies in the trade-off between pore volume and electrode density / conductivity. Excessive activation to create a high surface area will destroy the conductive network, and excessive doping may introduce unstable surface functional groups, thereby damaging the cycle life.

Overall, MCMBs exhibit distinct functional roles in different electrochemical systems, and their structural regulation based on specific applications is a key advantage, which makes MCMBs a multifunctional carbon material with broad prospects in the field of advanced energy storage. However, to exploit this potential, it depends on whether the academia and industry can harness the inherent trade-offs in materials science and develop scalable, cost-effective synthetic routes that can be tailored to meet the unique needs of each application.

## Conflicts of interest

The authors declare that they have no conflict of interest.

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