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Conjugated microporous polymer electrodes for supercapacitors: recent progress, key challenges, and future directions

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Conjugated Microporous Polymers (CMPs) have shown great potential as a class of materials for supercapacitor electrodes, offering a distinctive blend of extensive surface area, tunable porosity, redox activity, and chemical stability. This review thoroughly explores the role of CMPs in advancing supercapacitor technology, focusing on their structural and chemical characteristics, energy storage mechanisms, and recent advancements in material design and device engineering. We highlight the synergistic integration of CMPs with carbon-based nanomaterials, metal oxides, and conductive polymers to create hybrid and composite systems that enhance conductivity and electrochemical performance. Recent studies demonstrate significant improvements in essential performance indicators, including mass-based capacitance, energy storage capacity, power output efficiency, and cycling stability, positioning CMPs as competitive alternatives to traditional carbon-based materials like activated carbon as well as graphene. Although progress has been made, issues with scalability, conductivity, and long-term stability are still major challenges, requiring further research and innovation. This review also explores future directions, emphasizing the potential of CMPs in flexible, wearable, and solid-state supercapacitors, as well as their integration into hybrid energy systems. By addressing current limitations and leveraging emerging trends, CMP-based supercapacitors hold immense promise for enabling the next generation of highly durable, sustainable, and efficient energy storage systems. This review seeks to inspire future research as well as collaboration in this sector, facilitating transformative advances in energy storage systems.

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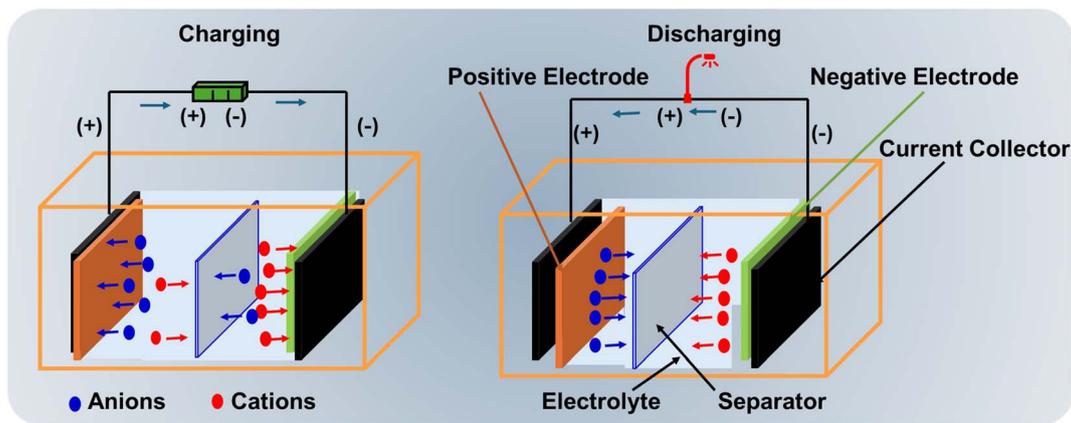
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Scheme 1 Illustration of the charge and discharge mechanisms in supercapacitors, highlighting ionic movement within the electrolyte and electron transport through the outer circuit.⁹

1. Introduction

The rapid growth of renewable energy sources, portable electronics, and electric vehicles has intensified the growing interest in advanced energy storage technologies capable of providing high power and energy densities, extended cycle life, and rapid charge–discharge performance.^{1–3} Among different energy storage systems, supercapacitors (or electrochemical capacitors) have become essential devices, bridging the performance gap between conventional capacitors and batteries.^{4–8} Real-world supercapacitors are generally composed of six essential components: a positive electrode, a negative electrode, an electrolyte, a separator, current collectors, and an outer encapsulation layer. During charge and discharge cycles, energy is stored and released at the electrode interfaces, requiring the synchronized transport of ions within charge carriers in the electrolyte and electrons traveling *via* an external

circuit to preserve overall charge neutrality, as shown in Scheme 1.⁹

Supercapacitors store energy through either the build-up of electrostatic charge at the interface between electrodes and electrolyte, involving electrical double-layer capacitance characterized by non-faradaic processes, commonly referred to as EDLC (Scheme 2a) or reversible faradaic reactions (pseudocapacitance) (Scheme 2b), enabling them to deliver high power densities and exceptional cycling retention.^{9–13} However, the relatively low energy density of supercapacitors compared to batteries remains a significant challenge, driving the search for innovative, economical, and sustainable electrode materials that can enhance their performance.^{14–16}

The presence or absence of distinct peaks in CV curves serves as a key indicator distinguishing non-faradaic charge storage in EDLCs from faradaic redox-driven processes in pseudocapacitive and battery-type materials, as shown in Scheme 3.

In recent years, conjugated microporous polymers (CMPs) have garnered considerable interest as a promising class of polymeric materials for supercapacitor electrodes.^{17–22} CMPs, a subset of porous organic polymers, are distinguished by their extended π -conjugated backbones and permanent microporosity.^{23–29} These materials uniquely integrate high surface area, tuneable pore structures, and excellent chemical stability with the electronic functionalities of conjugated systems, rendering them well-suited for use in energy storage. Unlike traditional carbon-based materials, which primarily rely on EDLC, CMPs can exhibit both EDLC and pseudocapacitance, offering the potential for higher energy densities without compromising power density or cycling stability.^{21,30,31}

Actually, the structural and chemical versatility of CMPs allows for precise control over their properties, such as porosity, conductivity, and redox activity, through rational design and synthesis.^{32–35} For instance, the incorporation of redox-active functional groups or heteroatoms into the CMP framework can introduce chemical pseudocapacitive behaviour, while the integration of conductive additives (*e.g.*, graphene and carbon nanotubes) can enhance physical electrical conductivity.^{36–41} Furthermore, the lightweight and processable nature of CMPs

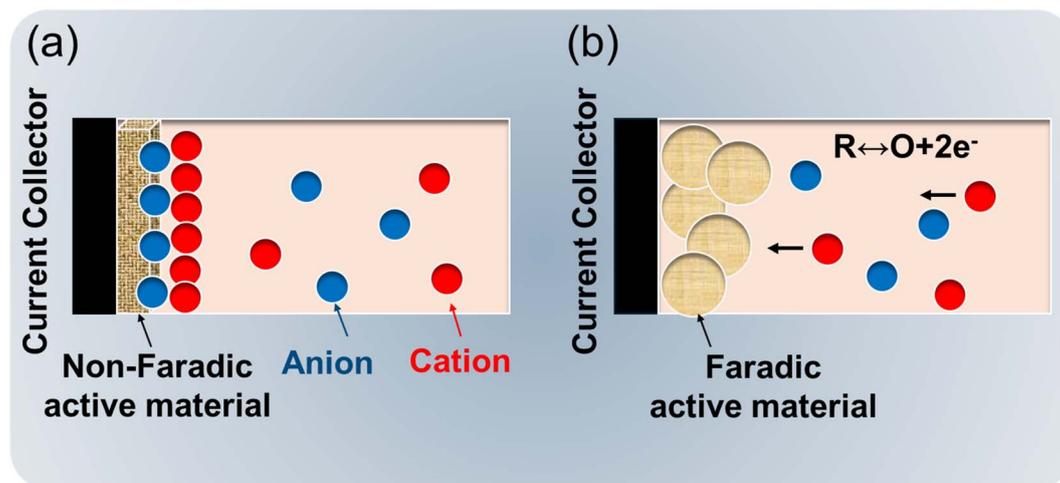


Shiao-Wei Kuo

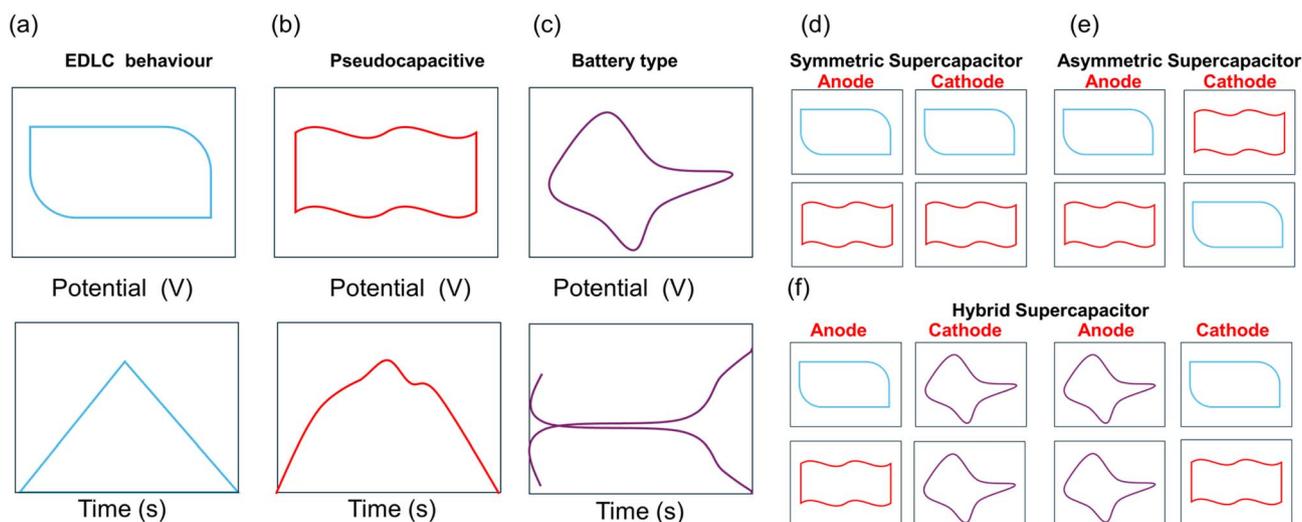
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Scheme 2 Diagrammatic representations of (a) non-faradaic and (b) faradaic mechanisms of charge storage.



Scheme 3 Illustration of current–potential characteristics for EDLCs (a), pseudocapacitors (b), and battery-type systems (c), showing the contrast between potential-independent and potential-dependent charge storage, along with different types of supercapacitors—symmetric (d), asymmetric (e), and hybrid configurations (f).

render them appropriate for flexible and wearable energy storage systems, expanding their applicability beyond traditional supercapacitors.^{42,43} Despite their potential, the development of CMP-based supercapacitors faces several challenges, including limited intrinsic conductivity, scalability of synthesis, and long-term stability under harsh operating conditions. Addressing these challenges requires a multidisciplinary approach, combining advances in material design, synthetic chemistry, and device engineering. Our work focuses on delivering a broad and detailed overview of the role of CMPs in supercapacitor development, highlighting their structural and chemical characteristics, energy storage mechanisms, recent advancements, and future opportunities. By exploring the unique advantages and limitations of CMPs, this review seeks to inspire further research and innovation in the field, opening the way to next-generation energy storage systems.

Despite the valuable insights provided by existing reviews on supercapacitors and CMPs, a dedicated review focusing on the role of CMPs in supercapacitor applications is both timely and essential. While prior studies have broadly discussed CMPs in contexts such as flexible electronics, catalysis, and batteries, there is a critical need for a comprehensive examination of their unique advantages and tailored advancements specifically for supercapacitor technologies.^{17,44–46} This review addresses this gap by providing a focused analysis of how CMPs—with their tuneable porosity, redox activity, and chemical stability—outperform traditional materials in supercapacitor applications, particularly under specific operational conditions. Furthermore, this review also integrates the latest developments and emerging trends in the field, including innovative hybrid systems, advanced configurations, and novel synthesis strategies that leverage CMPs to push the boundaries of energy



storage capabilities. Additionally, it critically identifies and addresses key research gaps, such as long-term stability, scalability in production, and economic feasibility, offering a roadmap for future investigations. By consolidating recent advancements and highlighting unexplored opportunities, this review aims to serve as a foundational resource for researchers and industry professionals, driving the next wave of innovation in supercapacitor technologies.

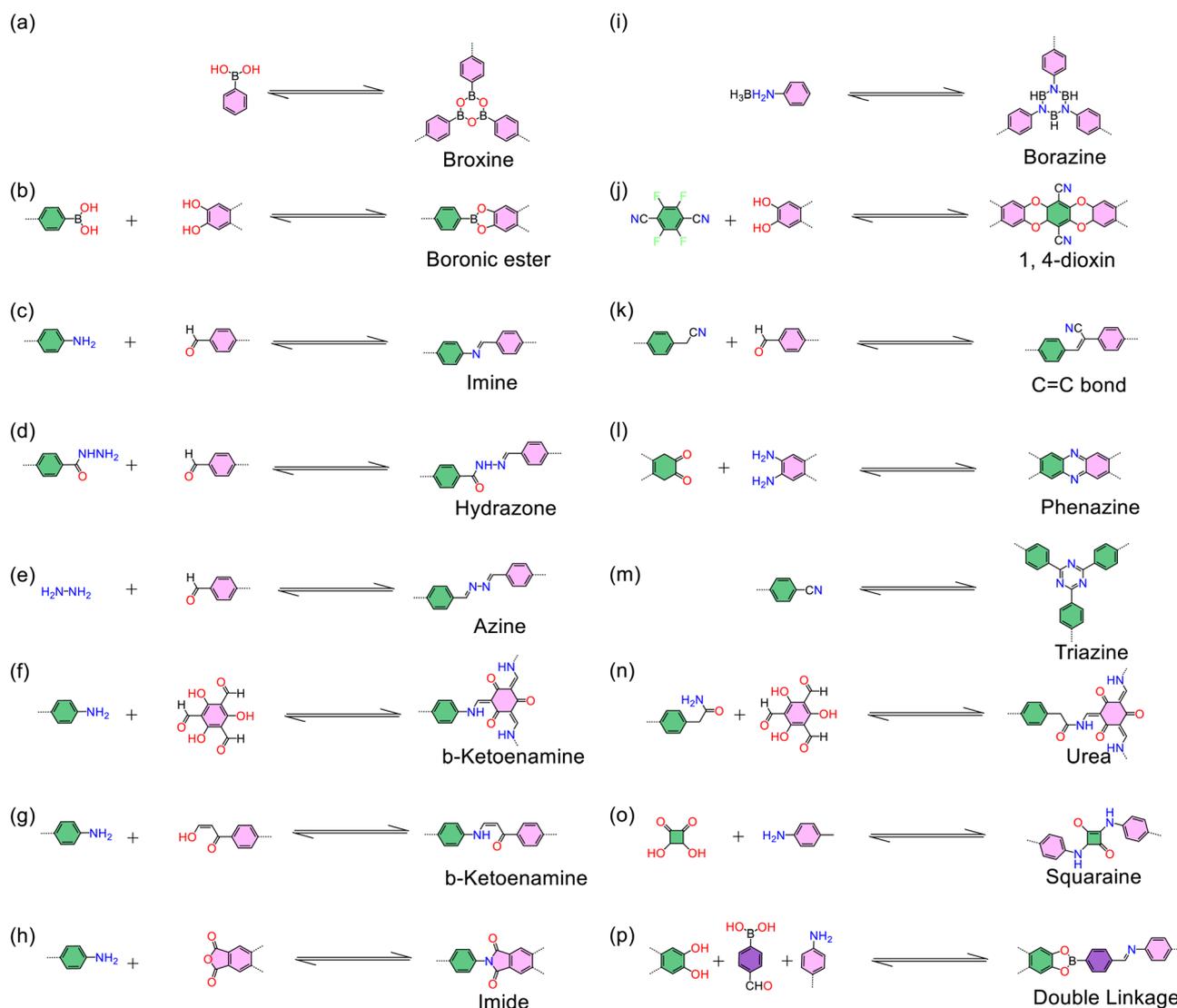
2. Structural and chemical characteristics of CMPs

CMPs represent a distinct category of porous organic materials that merge the benefits of extended π -extended conjugated networks possessing intrinsic microporosity (Scheme 4).^{46–49} These materials are characterized by the broad surface area they offer, tunable porosity, and chemical stability, making them highly attractive for energy storage technologies, particularly in

supercapacitors.^{50,51} The structural and chemical properties of CMPs are intrinsically linked to their molecular design, synthetic versatility, and the interplay between conjugation and porosity.^{52–54} This section delves into the architectural features of CMPs, their synthesis protocols, and the significance of their π -conjugated structures in enhancing supercapacitor performance.

2.1. Molecular architecture and design principles

CMPs are constructed from rigid, aromatic building blocks connected through covalent bonds, forming a highly cross-linked three-dimensional network. The design of CMPs is guided by the following principles, including rigid monomers, covalent linkages, and tunable functionality. In general, the use of rigid, planar aromatic monomers (*e.g.*, benzene, triazine, thiophene, and pyrene) ensures the formation of a stable and porous framework. These monomers provide structural rigidity, preventing the collapse of pores and maintaining the integrity



Scheme 4 Common types of covalent linkages formed through condensation reactions in COFs.⁶¹ Adapted with authorization of Elsevier.



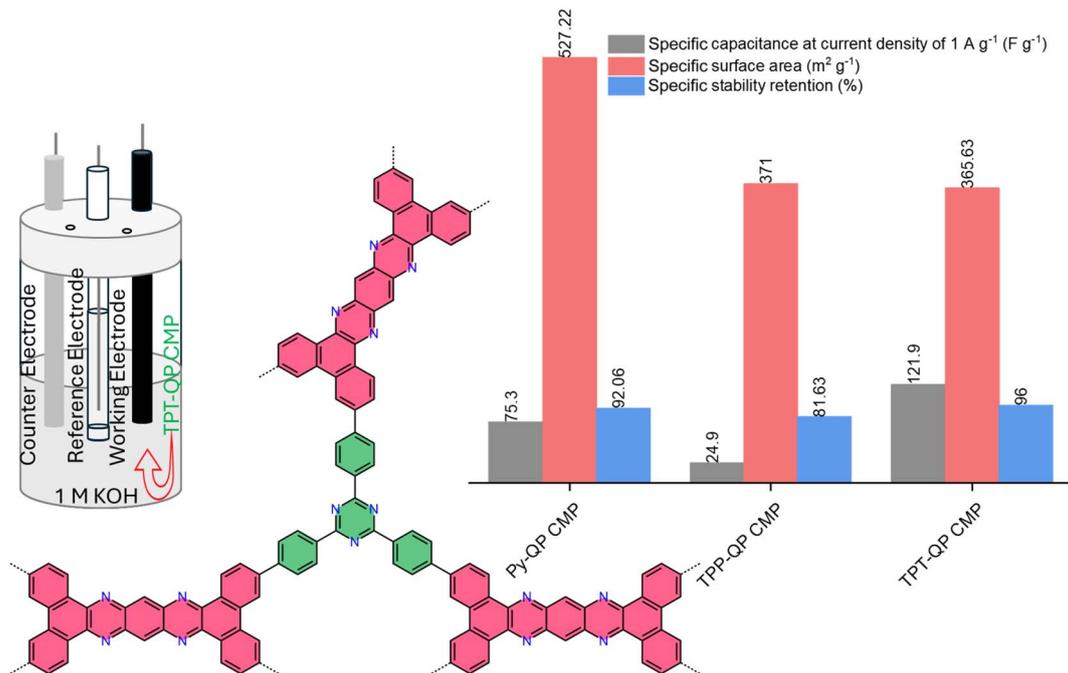


Fig. 1 Illustration showing different CMPs featuring a redox-active QP core and subunits such as Py, TPP, and TPT—with varying degrees of planarity and imine linkages—showing how these structural variations affect the physical and electrochemical properties when used as supercapacitor electrodes.²⁰ Adapted with authorization of Elsevier.

of the material under operational conditions.^{55–58} Our group reported the synthesis of three redox-active phenazine-based CMPs by coupling 3,6,14,17-tetrabromodibenzo[*a,c*]dibenzo [5,6:7,8]quinoxalino[2,3-*i*]phenazine (QP-4Br) with distinct boron-functionalized linkers: pyrene (Py), modified pyridine (TPP), and modified triazine (TPT).²⁰ These CMPs exhibit excellent thermal resistance, as indicated by char residue reaching up to 82.5 wt%, and enhanced surface areas reaching as high as 527 m² g⁻¹. Notably, the electrochemical performance of these materials is influenced by the planar configuration and imine linkage content of the subunits, which directly impact their energy storage behaviour. Among the synthesized CMPs, the most planar TPT-QP CMP demonstrates the most favourable structural characteristics, achieving an impressive specific capacitance of 121.9 F g⁻¹ and an energy density of 16.93 Wh kg⁻¹ as shown in Fig. 1.¹⁸ Additionally, it exhibits outstanding stable cycling behaviour, preserving 96% of capacitance after 5000 runs, making its performance comparable to other state-of-the-art porous electrode materials.

For covalent linkage, the monomers are connected through robust covalent bonds (*e.g.*, C–C, C–N, and C–O) formed *via* coupling reactions such as Suzuki, Sonogashira, or oxidative coupling throughout CMP frameworks. These linkages ensure the permanence of the porous structure and contribute to the chemical stability of CMPs as shown in Scheme 4.^{59,60}

For tuneable functionality, the incorporation of functional groups or heteroatoms into the CMP framework allows for the fine-tuning of properties such as wettability, conductivity, and redox activity.^{62,63} This tunability is also critical for optimizing CMPs for specific energy storage applications. Fig. 2 displays

a couple of CMPs exhibiting redox behaviour, TAT-CMP-1 and TAT-CMP-2, which were successfully designed and synthesized using nitrogen-dense, highly conductive triazatruxene molecule-based building subunits.⁶⁴

Despite their intermediate surface areas of 88 m² g⁻¹ for TAT-CMP-1 and 106 m² g⁻¹ for TAT-CMP-2, they demonstrated remarkable electrochemical performance, achieving specific capacitances of 141 F g⁻¹ and 183 F g⁻¹, respectively, at a current density of 1 A g⁻¹. Notably, both materials exhibited exceptionally specific capacitance values exceeding 160 μF cm⁻², which results from their redox-active nitrogen-rich structures that enhance pseudocapacitive behaviour. Furthermore, TAT-CMP-2 demonstrated outstanding cycling stability, retaining 95% compared to its starting capacitance after 10 000 charge–discharge runs at a high current density of 10 A g⁻¹. This superior durability highlights the prospects of these materials in long-term use in electrochemical energy storage devices.

2.2. Synthesis methods

The synthesis of CMPs involves a variety of chemical reactions and techniques, each providing distinct benefits in terms of precise control of the structure and functionality as displayed in Scheme 4. Noble metal-catalysed coupling reactions, such as Suzuki–Miyaura (Scheme 4a) and Sonogashira (Scheme 4b) couplings, are widely used to construct CMPs.^{65,66} These reactions enable the formation of C–C and C–N bonds, resulting in highly conjugated and porous networks. Coupling reactions are a cornerstone of recent synthetic chemistry, enabling the formation of covalent C–C or C–N bonds with high precision



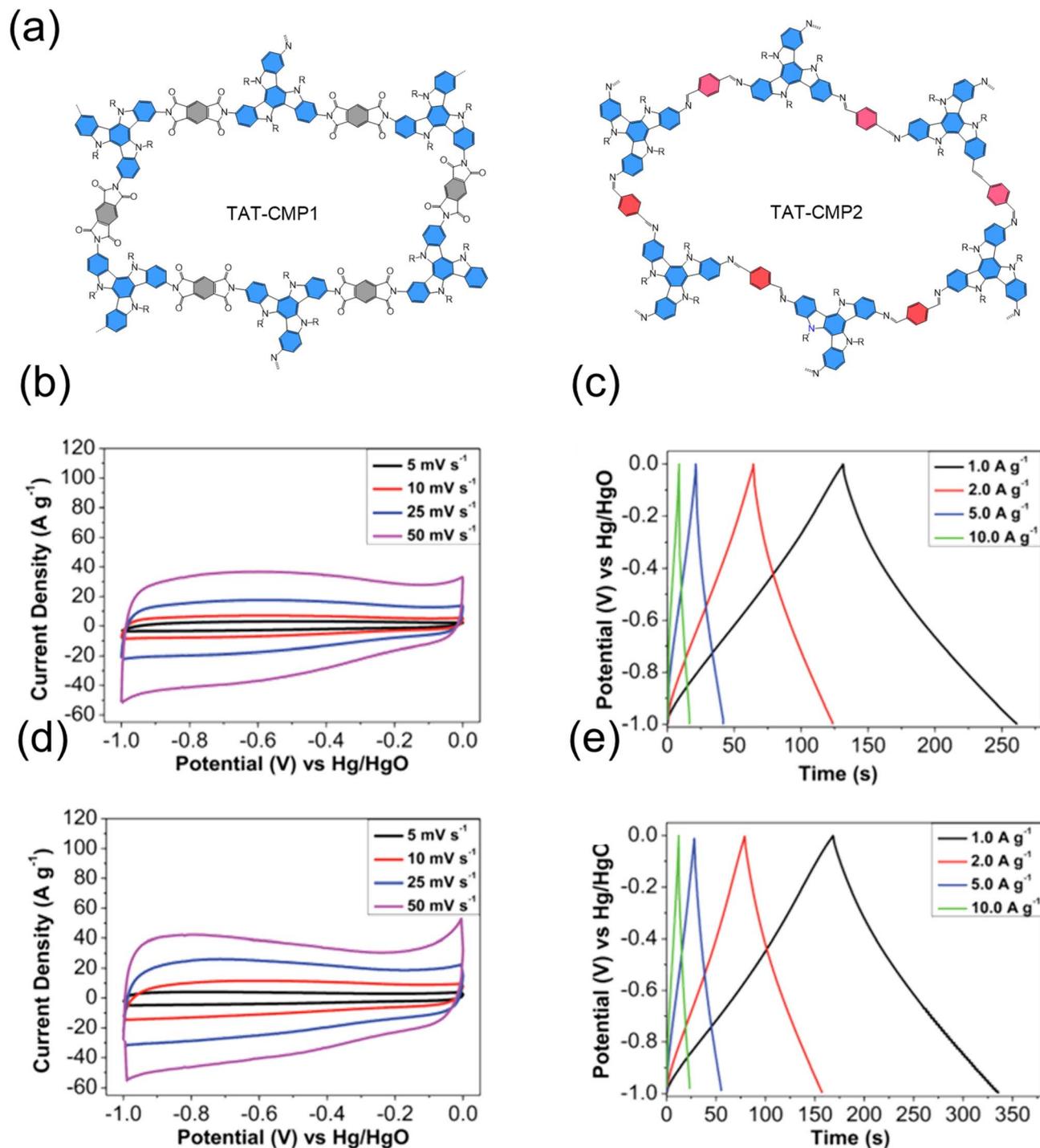


Fig. 2 (a) Schematic structures of TAT-CMP-1 and TAT-CMP-2, and (b and d) CV and (c and e) GCD analyses of TAT-CMP-1 (b and c) and TAT-CMP-2 (d and e), respectively.⁶⁴ Adapted with authorization of the Royal Society of Chemistry.

and efficiency. These reactions are critical for achieving the structural and electronic properties necessary for supercapacitor devices.

The Suzuki–Miyaura coupling is a palladium-catalyzed bond-forming reaction between an organoboron compound (boronic acid or boronic ester) and an organic halide (or pseudohalide) as displayed in Scheme 4a. This reaction is widely used in CMP

synthesis due to its versatility, mild reaction conditions, and compatibility with a wide range of functional groups.⁶⁷ Actually, the Suzuki–Miyaura coupling proceeds through a catalytic cycle involving three key steps. The palladium(0) catalyst inserts into the carbon–halide bond of the organic halide, forming a palladium(II) complex. Then, the organoboron compound transfers its organic group to the palladium(II) complex, facilitated by



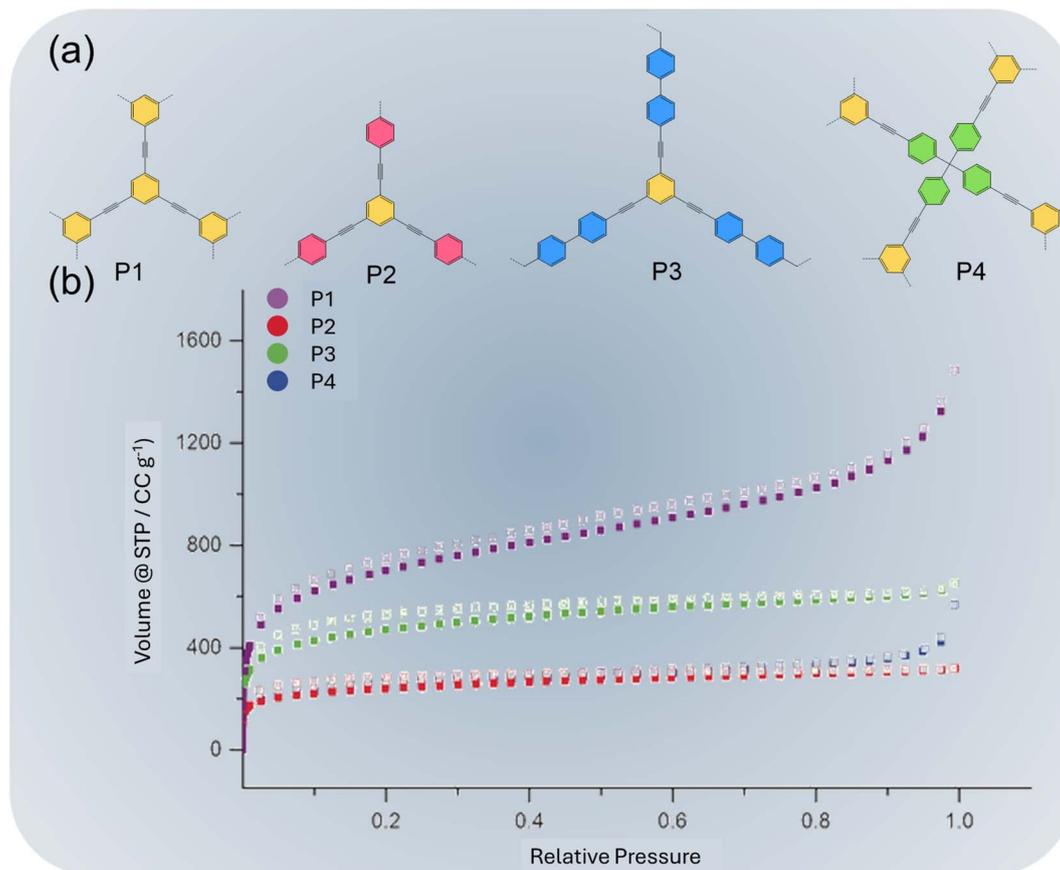


Fig. 3 (a) Copper-free CMPs synthesized from 1,3,5-triethynylbenzene and used as supercapacitor electrodes and (b) corresponding N_2 adsorption-desorption isotherms.⁷⁴ Adapted with authorization of Wiley.

a base (e.g., K_2CO_3). After that, the palladium(II) complex proceeds through reductive elimination to form the desired C–C bond, regenerating the palladium(0) catalyst.⁶⁸ These reactions often offer high functional group tolerance, allowing for the incorporation of diverse monomers into CMPs. The mild reaction conditions (typically room temperature to 110 °C) reduce the risk of side reactions.⁶⁹ Moreover, compatibility with aqueous or mixed solvent systems makes it environmentally friendly. Realistically, the Suzuki–Miyaura coupling has been extensively used to design CMPs possessing elongated π -conjugation as well as large surface areas. Within our earlier work, we designed a Py based CMP through the classical Suzuki protocol, which shows a high surface area of up to 1470 $m^2 g^{-1}$ and an excellent specific capacitance as an organic supercapacitor electrode.⁷⁰ The reaction's versatility enables the incorporation of redox-active functional groups (e.g., quinones and thiols) and heteroatoms (e.g., nitrogen and sulfur), further enhancing pseudocapacitance.^{21,71}

On the other hand, the Sonogashira coupling is a palladium- and copper-catalyzed reaction between a terminal alkyne and an aromatic or vinyl halide (or pseudohalide), as displayed in Scheme 4b.⁵⁴ The Sonogashira coupling is particularly valuable for constructing CMPs with alkyne linkages, which contribute to extended π -conjugation and structural rigidity. It proceeds

through a catalytic cycle involving insertion of a palladium(0) catalyst into the carbon–halide bond of the organic halide, forming a palladium(II) complex. Then, the terminal alkyne reacts with a copper(I) catalyst under basic conditions (e.g., triethylamine) to form a copper acetylide intermediate. The copper acetylide transfers the alkyne group to the palladium(II) complex. Ultimately, the palladium(II) complex goes through reductive elimination to form the desired C–C bond, reproducing the palladium(0) catalyst. Realistically, Sonogashira coupling enables the formation of C \equiv C (alkynes), which enhances π -conjugation and electronic conductivity.⁷² Furthermore, it offers high selectivity and efficiency, even in the presence of sensitive functional groups. Furthermore, it is compatible with a wide range of solvents, including polar and non-polar systems. As a result, Sonogashira coupling has been widely employed to synthesize CMPs with alkyne linkages, which impart structural rigidity and high surface areas.⁷³ For example, Trunk *et al.* proposed a copper-free, single-step Sonogashira cross-coupling reaction that successfully produced structurally precise conjugated microporous poly(aryleneethynylene) networks as shown in Fig. 3 with unparalleled surface areas reaching 2552 $m^2 g^{-1}$, establishing a new benchmark for poly(aryleneethynylene) networks and



demonstrating their potential for advanced energy storage applications.⁷⁴

The reaction's ability to incorporate alkyne groups also allows for post-synthetic modifications, such as click chemistry, to introduce additional functionalities.⁷⁵ Clearly, such a combination of Suzuki–Miyaura and Sonogashira couplings in CMP synthesis enables the creation of complex, multifunctional networks with tailored properties. For example, a CMP synthesized using both coupling reactions can incorporate both aryl-aryl and aryl-alkyne linkages, resulting in a highly conjugated and porous framework with enhanced electrochemical performance. This synergistic approach enables accurate regulation of the structural and electronic properties of CMPs, optimizing them for specific applications such as supercapacitors. Imine-linked CMPs were synthesized through Schiff base reactions, as shown in Scheme 4c, and offer excellent crystallinity and stability.⁷⁶ Schiff base chemistry, characterized by the formation of imine (C=N) bonds through the reaction between aldehydes (or ketones) and primary amines, has emerged as a powerful tool for constructing CMPs. Imine-linked CMPs, synthesized *via* Schiff base reactions, exhibit excellent crystallinity, chemical stability, and tuneable functionality. The reversible nature of imine bonds further enables post-synthetic modifications, making these materials highly versatile for applications in energy storage, catalysis, and gas separation.⁷⁷ Furthermore, this reaction proceeds under mild conditions, typically under acid-catalysed conditions or at elevated temperatures. Ruidas *et al.*⁷⁸ reported that imine bonds in the synthesized π -conjugated COFs, namely TFPh-NDA and TFR-NDA, as displayed in Fig. 4, which can play a pivotal role in enhancing structural stability, redox activity, and electrochemical performance, enabling exceptional gravimetric charge storage capacity, energy density, and durability during cycling, thereby highlighting their critical importance in advancing high-

performance supercapacitor materials for energy storage applications.

From a mechanistic point of view, the primary amine serves as a nucleophile, interacting with the electrophilic carbon centre of the aldehyde or ketone, forming a hemiaminal intermediate. Then, the hemiaminal intermediate undergoes dehydration, facilitated by the removal of water, to form the imine bond. Importantly, the imine bond is dynamic and reversible, allowing for the exchange of components under specific conditions (*e.g.*, acidic environment). Again, this reversibility is a key feature that enables post-synthetic modifications and self-healing features in imine-linked CMPs. Schiff base reactions offer several advantages for the synthesis of CMPs, making them a popular choice for constructing porous and functional materials. As mentioned above, imine-linked CMPs often exhibit high crystallinity due to the directional nature of the Schiff base reaction, which promotes the formation of well-ordered structures as well as sometimes COFs, as shown in Scheme 4. Realistically, this crystallinity enhances the material's stability and performance in applications such as supercapacitors. Further, imine bonds are chemically stable under a wide range of conditions, rendering imine-linked CMPs suitable for use in harsh environments, including moderately acidic, basic, and oxidative conditions. Furthermore, the choice of aldehyde and amine monomers offers precise adjustment of the chemical as well as structural properties of the resulting CMPs. Importantly, functional groups (*e.g.*, -OH, -NH₂, and -COOH) can be incorporated into the monomers to introduce additional reactivity or binding sites.⁷⁹

Imine-linked CMPs are broadly explored for various applications, particularly in energy storage, catalysis, and gas separation. These properties enable their effective use in supercapacitor electrodes, where high surface area, redox activity, and stability are critical. Moreover, imine-linked CMPs

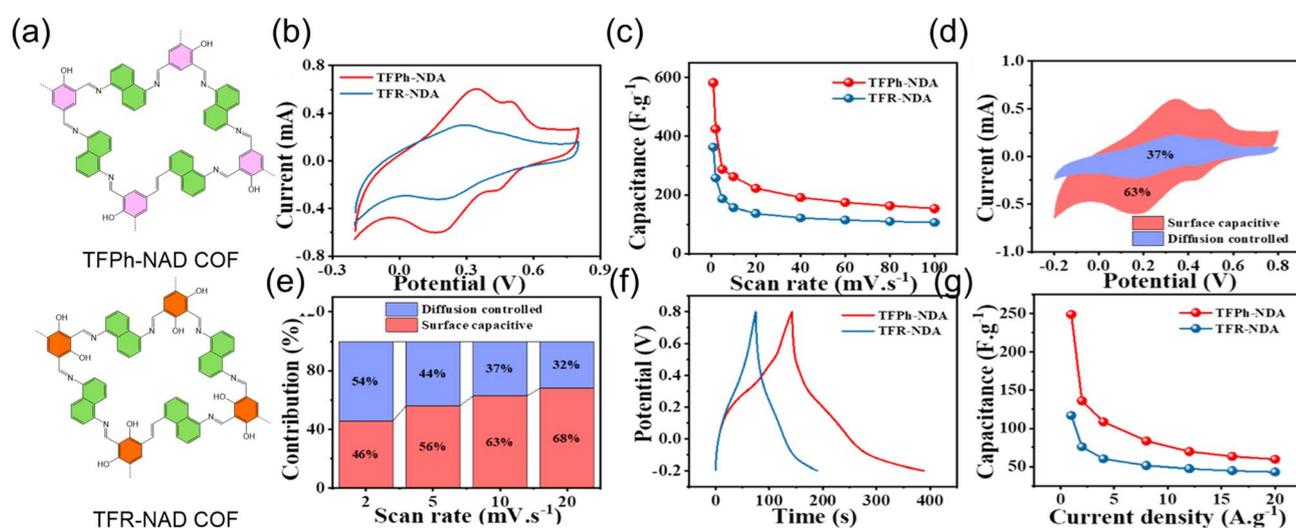


Fig. 4 (a) Chemical structures of TFPh-NDA and TFR-NDA COFs, (b) comparison of their CV curves at 10 mV s⁻¹ in a three-electrode setup, (c) specific capacitance *versus* scan rate from CV data for both materials, (d) contribution of surface-controlled and diffusion-controlled capacitance for TFPh-NDA in 1 M H₂SO₄ at 10 mV s⁻¹, (e) bar chart showing these contributions at different scan rates for TFPh-NDA COF, (f) comparison of GCD curves at 1 A g⁻¹, and (g) specific capacitance *versus* current density based on GCD results for TFPh-NDA and TFR-NDA COFs.⁷⁸ Adapted with authorization of American Chemical Society.



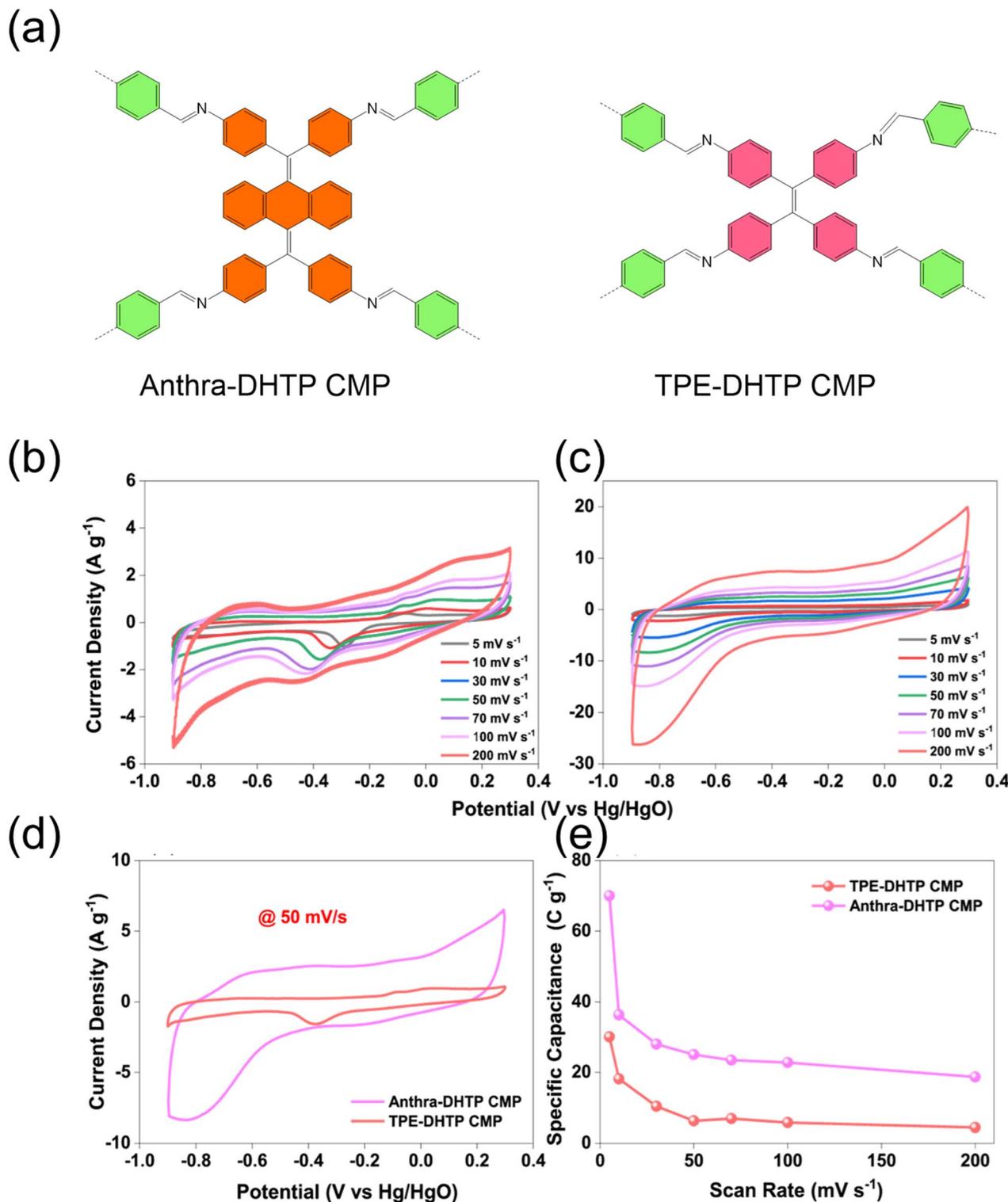


Fig. 5 (a) Chemical structures of TPE-DHTP and Anthra-DHTP CMPs, (b and c) CV analyses of TPE-DHTP and Anthra-DHTP CMPs, (d) representative CV curves of DHTP-based CMP electrodes measured at 50 mV s^{-1} , and (e) plot illustrating the dependence of specific capacitance on the scan rate for the DHTP-CMP electrodes.⁸⁰ Adapted with authorization of American Chemical Society.

exhibit redox-active sites, rendering them ideal for energy storage systems. We proved that inclusion of imine bonds in the Anthra-DHTP CMPE as shown in Fig. 5 significantly enhances its electrochemical performance as a supercapacitor material by

providing structural stability, redox-active sites, and improved charge transfer capabilities, which contribute to its remarkable specific capacitance of 121 F g^{-1} at 0.5 A g^{-1} and a high capacitance cycling rate of 79% over 5000 cycles.⁸⁰



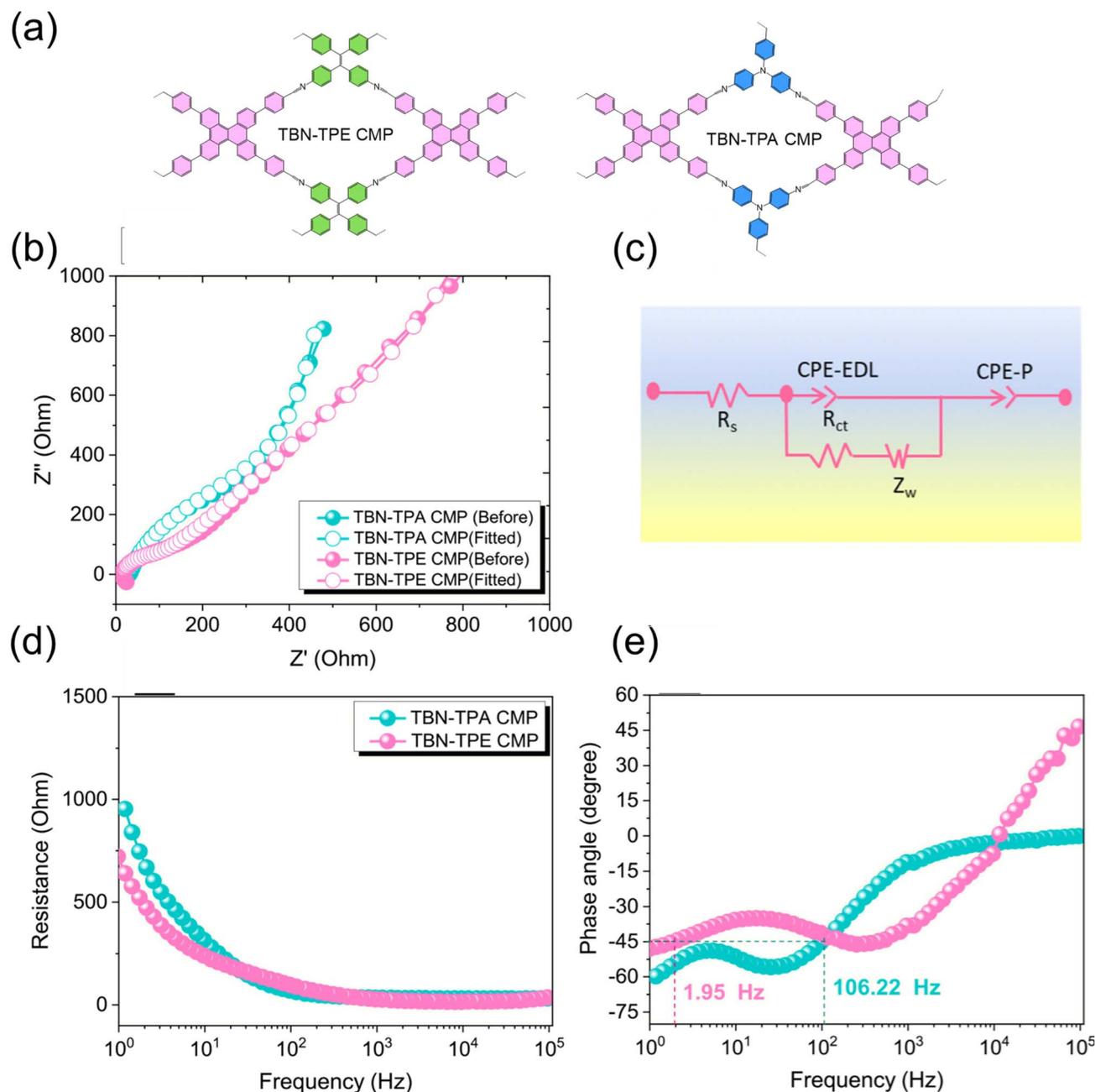


Fig. 6 (a) Molecular structures of TBN-TPE and TBN-TPA CMPs, (b) Nyquist plots illustrating the electrochemical impedance behaviour of both materials, (c) corresponding equivalent circuit models used for data fitting, (d) Bode plots showing the relationship between frequency and impedance magnitude, and (e) Bode phase plots displaying frequency versus phase angle to identify the knee frequency.⁸¹ Adapted with authorization of Elsevier.

The dynamic imine bonds enable the exchange of monomers within the CMP framework, allowing for the introduction of new functional groups or the optimization of pore sizes as represented through those designed CMPs, namely TBN-TPE and TBN-TPA CMPs, as shown in Fig. 6.⁸¹ Moreover, imine-linked CMPs can be functionalized with additional groups (*e.g.*, redox-active moieties and metal complexes) through post-synthetic reactions, enhancing their electrochemical features.

While Schiff base chemistry offers numerous advantages for CMP synthesis, several challenges still remain. For example,

those imine bonds can hydrolyse under strongly acidic or basic conditions, limiting the stability of imine-linked CMPs in aqueous electrolytes.^{77,82} Developing strategies to enhance hydrolytic stability is an ongoing area of research. Scaling up the synthesis of imine-linked CMPs while maintaining crystallinity and functionality requires optimization of reaction conditions and monomer availability. In addition, some aldehyde and amine monomers used in Schiff base reactions can be expensive or difficult to synthesize, impacting the cost-effectiveness of imine-linked CMPs. Therefore, we expect that



upcoming studies should aim to overcome these challenges through the development of novel monomers, advanced characterization techniques, and innovative synthetic strategies. The continued exploration of imine-linked CMPs holds great promise for advancing materials science and enabling next-generation technologies.

The oxidation polymerization involves the oxidative coupling of aromatic monomers, resulting in the formation of conjugated polymers with inherent porosity. Oxidative polymerization is particularly useful for synthesizing CMPs with extended π -conjugation.⁸³ This method involves the oxidative coupling of aromatic monomers, typically catalysed by transition metal salts or chemical oxidants, to form highly conjugated polymer networks. The resulting CMPs exhibit excellent electronic properties, expansive surface areas, and customizable pore networks; they are well-suited for supercapacitor applications.²⁹ Gu *et al.* stated that the electrooxidative polymerization protocol of CMP films enables precise control over film thickness, combining significant porosity, delocalized π -electron systems, and fast electron transport.⁸⁴ Technically, oxidative polymerization proceeds through the formation of radical intermediates, which couple to form covalent bonds between aromatic monomers. The reaction is typically initiated by an oxidant, such as FeCl_3 or AlCl_3 , which abstracts electrons from the monomers to generate radical cations.⁸⁵ These radical cations then undergo coupling reactions to form the polymer network. On the other hand, the oxidant abstracts an electron from the aromatic monomer, generating a radical cation intermediate. Then, the radical cations couple together, forming covalent bonds between those reacted monomers and extending the π -conjugated framework. After that, the coupled intermediates lose protons to restore aromaticity, resulting in the formation of a stable conjugated polymer. Therefore, the rigid and planar nature of the aromatic monomers, combined with the random coupling process, leads to the formation of a microporous structure. The porosity is inherent to the polymer network and does not require additional templating agents. Realistically, oxidative polymerization offers several advantages for the synthesis of CMPs, making it a popular choice for constructing materials with tailored electronic and structural properties. The oxidative coupling of aromatic monomers results in highly conjugated polymer networks, which facilitate efficient charge transport and enhance electronic conductivity.⁸⁶ This property is particularly beneficial for supercapacitor electrodes, where rapid charge-discharge cycles are required. Furthermore, the random coupling of rigid aromatic monomers creates a microporous structure with high surface areas.⁸⁷ This inherent porosity provides abundant active sites for ion adsorption and electrolyte interaction, contributing to high capacitance. Furthermore, oxidative polymerization is a straightforward and scalable method that does not require complex catalysts or reaction conditions. It can be performed either dissolved or as a solid, making it suitable for wide production. Therefore, oxidative polymerization has been utilized to design CMPs for various applications, particularly in energy storage.⁸⁸ Its ability to create highly conjugated and porous materials makes it particularly suitable for supercapacitor electrodes.

Furthermore, CMPs synthesized *via* oxidative polymerization can exhibit high surface areas and redox-active sites, rendering them ideal for energy storage systems.⁸⁹ While oxidative polymerization offers numerous advantages for CMP synthesis, several challenges remain. The random nature of oxidative coupling can lead to irregular pore sizes and distributions, which may hinder ion transport and accessibility.⁹⁰ Developing strategies to control pore structures is an ongoing area of research. Moreover, the choice of oxidant can significantly impact the reaction efficiency and the properties of the resulting CMPs. Some oxidants (such as FeCl_3) can be difficult to remove from the final product, requiring additional purification steps. Notably, the use of chemical oxidants can raise environmental and safety concerns. Developing greener oxidants or alternative methods for oxidative polymerization is a priority for sustainable production. Therefore, future studies should focus on addressing these challenges through the development of novel monomers, advanced characterization techniques, and innovative synthetic strategies. The continued exploration of oxidative polymerization holds great promise for advancing materials science and enabling next-generation technologies.

On the other scale, the template-assisted synthesis is a sophisticated strategy for constructing CMPs with hierarchical porosity. Actually, template-assisted synthesis involves the utilization of a sacrificial template to create a predefined pore structure within the CMP framework.⁹¹ The template is removed after polymerization, leaving behind a porous network with controlled pore sizes and distributions. The process can be categorized into two main approaches: soft templates, such as surfactants or block copolymers, self-assemble into micelles or other nanostructures during polymerization. These nanostructures act as placeholders, guiding the formation of pores in the CMP framework. After polymerization, the soft templates are removed through solvent extraction or thermal decomposition. On the other hand, hard templates, such as silica nanoparticles, colloidal crystals, or anodic aluminium oxide (AAO), provide a rigid scaffold for polymerization. The CMP forms around the template, and the template is subsequently removed through chemical etching or calcination, leaving behind a porous structure. Zuo *et al.* utilized a template-assisted method through NH_2 -MWNTs (Fig. 7) for the design of core-shell PTPA@MWNTs, which significantly enhances the porous structure, redox behaviour, and electrical conductivity of CMPs, resulting in a remarkable specific capacitance of 410 F g^{-1} and good cycling stability (71% over 6000 cycles), demonstrating the effectiveness of this approach in optimizing CMPs designed for advanced electrochemical energy storage applications.⁹²

The use of templates enables the creation of CMPs with hierarchical porosity, combining micro-, meso-, and macropores. This hierarchical structure enhances ion transport and electrolyte accessibility, leading to enhanced electrochemical performance in supercapacitors. Moreover, templates allow for precise control over pore sizes and distributions, enabling the optimization of CMPs for specific applications. For example, mesopores can facilitate ion diffusion, while micropores provide a high surface area for ion adsorption. Template-assisted synthesis often results in CMPs with high surface



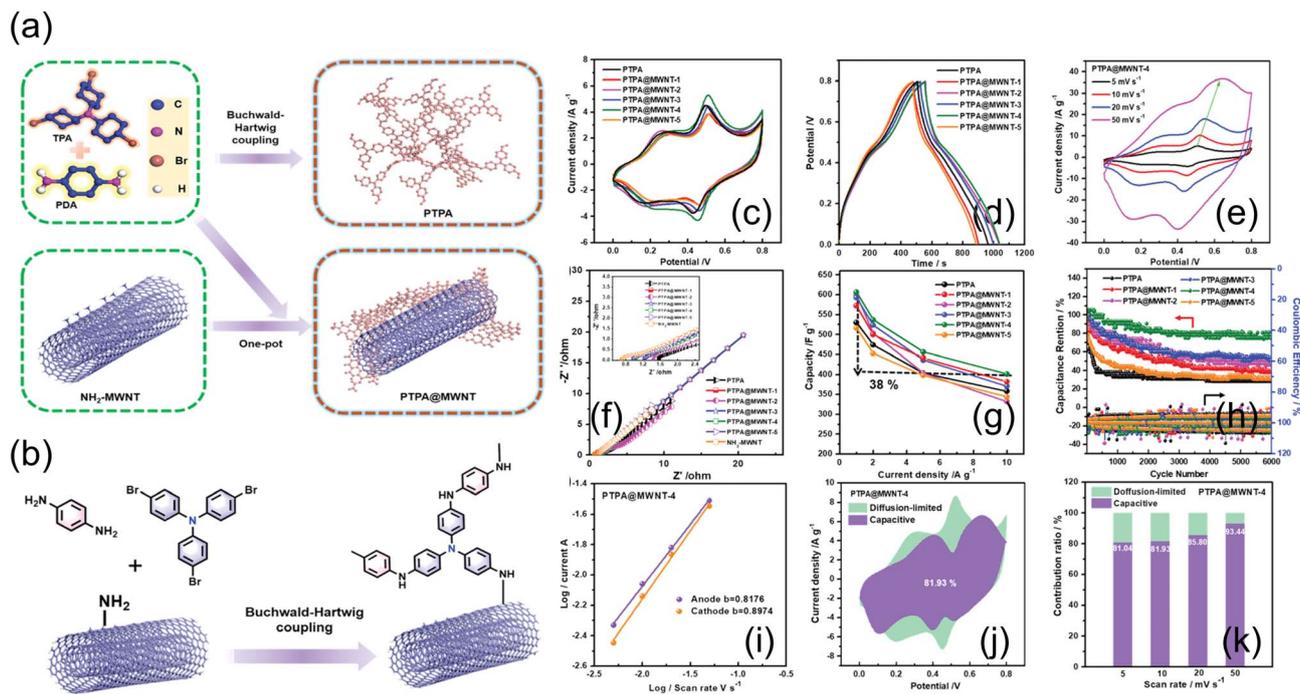


Fig. 7 (a) Synthetic route for PTPA@MWNT, (b) proposed structural model showing PTPA chains grafted onto NH_2 -functionalized MWNTs, (c) CV and (d) GCD curves of PTPA@MWNT composites (samples 1–5) measured at 1 A g^{-1} , (e) additional CV profiles and (f) Nyquist plots fitted with the equivalent circuit model with the inset highlighting the high-frequency region. (g) Specific capacitance values and (h) cycling stability of PTPA@MWNT-1 to -5 and pristine PTPA; (i) log–log plot of peak current (*i*) versus scan rate (ν) with linear fits; (j) deconvolution of capacitive and diffusion-controlled current contributions; and (k) quantitative analysis of capacitance versus diffusion-controlled charge storage contributions for PTPA@MWNT-4.⁹² Adapted with authorization of Wiley.

areas, providing abundant active sites for ion adsorption and hence enhanced energy storage.⁹¹ Although the template-assisted methodology shows numerous advantages for CMP synthesis, several challenges still remain; for example, the removal of templates, especially hard templates, can be challenging and may require harsh conditions such as strong acids or even high temperatures. Therefore, developing milder and more efficient template removal protocols is an ongoing area of research. In addition, scaling up template-assisted synthesis while maintaining control over pore structure and uniformity requires optimization of reaction conditions and template availability. Importantly, the use of templates can increase the cost and complexity of CMP synthesis. Developing cost-effective and readily available templates for CMP synthesis is crucial for mass production. We suggest that further investigation is needed to overcome these challenges through the development of novel templates, advanced characterization techniques, and innovative synthetic strategies. The continued exploration of template-assisted synthesis holds great promise for advancing materials science and enabling next-generation technologies.

2.3. π -conjugated structures

The extended π -conjugation in CMPs is a defining feature that distinguishes them from other porous families.⁹³ This conjugation arises from the overlap of π -orbitals in the incorporated aromatic units, creating a delocalized electron system that spans the polymer backbone.²⁴ The π -conjugated framework

not only imparts unique electronic properties to CMPs but also plays a critical role in their performance as supercapacitor electrodes.⁹⁴ The delocalized π -electrons in CMPs enable efficient charge transport along the polymer backbone, significantly enhancing the material's electrical conductivity.⁹⁴ This property is particularly advantageous for supercapacitor applications, where rapid electron transfer is key to reaching high power density. Furthermore, in π -conjugated systems, the overlap of π -orbitals creates a continuous pathway for electron delocalization.⁹⁵ This delocalization reduces the energy barrier for electron movement, allowing for rapid charge transport. The extended conjugation also minimizes charge trapping and recombination, further improving conductivity. Furthermore, the high conductivity of π -conjugated CMPs reduces the internal resistance of supercapacitors, enabling faster charge-discharge cycles and higher power densities.⁹⁶

The π -conjugated framework in CMPs can participate in redox reactions, contributing to pseudocapacitance. This redox activity is a key factor in achieving high energy densities in supercapacitors, as it allows for additional charge storage beyond the EDLC. Therefore, the π -conjugated system can undergo reversible oxidation and reduction reactions, often facilitated by the presence of heteroatoms (*e.g.*, nitrogen and sulfur) or functional groups (*e.g.*, quinones and thiols). These redox reactions involve the transfer of electrons and ions, leading to faradaic charge storage. The redox-active triazatruxene CMPs (TAT-CMP-1 and TAT-CMP-2) (Fig. 2) leverage



their nitrogen-rich structures to deliver exceptional pseudocapacitance, achieving exceptional specific capacitances of 141 F g^{-1} and 183 F g^{-1} , respectively, along with outstanding cycling stability (95% retention after 10 000 cycles), demonstrating the critical role of redox activity in improving the efficiency of CMP-derived supercapacitor electrodes.⁶⁴ Therefore, such a combination of EDLC and pseudocapacitance in π -conjugated CMPs results in higher specific capacitances and energy densities.^{20,97} Importantly, the performance of π -conjugated CMPs can be further enhanced through rational design strategies. Such a choice of aromatic monomers (*e.g.*, benzene, thiophene, and pyrene) determines the extent of π -conjugation and the electronic properties of the resulting CMPs.^{27,70,98} Planar and rigid monomers are preferred for maximizing conjugation. Moreover, the introduction of heteroatoms into the π -conjugated framework can enhance redox activity and conductivity. Therefore, the addition of functional groups to the π -conjugated backbone can introduce additional redox-active sites, further enhancing energy storage capabilities.

Despite the extended π -conjugation of CMPs, their conductivity is often lower than that of inorganic materials. Developing strategies to further enhance conductivity, such as hybridization with conductive materials or doping, is an ongoing area of research. Notably, the long-term stability of π -conjugated CMPs under harsh electrochemical conditions (*e.g.*, high voltage and extreme temperatures) requires further investigation. Ensuring structural integrity and performance over extended cycles is critical for real-world applications. Scaling up the synthesis of π -conjugated CMPs while maintaining control over their structural and electronic properties is a key challenge. Therefore, we believe that future studies need to concentrate on overcoming these challenges through advanced material design, innovative synthesis techniques, and comprehensive characterization studies. The continued exploration of π -conjugated CMPs holds great promise for advancing energy storage technologies.

2.4. Porosity and surface area

The pore structure and surface area of CMPs are critical factors that determine their performance in supercapacitor applications. The microporous structure of CMPs arises from the rigid, three-dimensional network formed during polymerization, which creates a density of pores and a large internal surface area. These features provide numerous active sites available for ion adsorption as well as electrolyte interaction, enabling efficient charge storage and transport. CMPs are well-known for their extremely large surface areas, which are a direct result of their microporous structure.⁹⁸ This high surface area is a key advantage for supercapacitor applications, as it provides numerous active sites for ion adsorption and electrolyte interaction. The wide surface area of these CMPs allows for such formation of a large electrical bilayer structure at the interface between electrodes and electrolyte (Scheme 2a), which is the primary mechanism of charge storage in EDLCs. The greater the surface area, the higher the specific capacitance, as more ions can be accumulated onto the electrode interface.⁵⁴ Surface area

is typically measured using gas adsorption techniques, such as nitrogen adsorption–desorption isotherms, and calculated using the BET protocol. Optimizing the surface area of CMPs involves selecting monomers with rigid and planar structures, as well as controlling polymerization conditions to maximize pore formation.⁹⁹

The pore size distribution throughout CMPs is a crucial factor that influences ion accessibility, electrolyte diffusion, and overall electrochemical performance. Pore sizes in CMPs are typically in the micropore range, although mesopores and macropores can also be present, depending on the synthesis method and monomer selection. The pore sizes of CMPs are customizable based on the choice of monomers with different geometries and sizes.^{21,98} Monomers such as pyrene or triphenylene tend to create wider pores due to their bulkier structures. Otherwise, monomers such as benzene or thiophene yield narrower pores, which are ideal for accommodating solvated ions and maximizing ion adsorption.¹⁰⁰ Therefore, optimal pore sizes are essential for matching the size of solvated ions in the electrolyte, ensuring efficient ion accessibility and maximizing capacitance. Pores that are too small may restrict ion diffusion, while pores that are too large may reduce the effective surface area available for ion adsorption.

Some CMPs exhibit hierarchical porosity, combining micro-, meso-, and macropores within a single material. This hierarchical structure enhances mass transfer and diffusion of ions, leading to improved rate capability as well as overall performance in supercapacitors.^{43,101} Micropores provide a high surface area for ion adsorption, contributing to high capacitance. Mesopores facilitate ion transport and electrolyte diffusion, reducing resistance and improving rate capability. On the other hand, macropores serve as ion reservoirs and transport pathways, further enhancing mass transport. Hierarchical porosity can be achieved through template-assisted synthesis, where soft or hard templates guide the formation of pores at multiple length scales.¹⁰² CMPs with hierarchical porosity exhibit enhanced electrochemical performance, including higher capacitance, superior rate performance, and improved long-term stability.¹⁷ Zhao *et al.*¹⁰⁰ reported that the exceptional surface area and hierarchical porous nature of pyrene-based CMP (SDBPy)-derived carbon, optimized through KOH activation, enable high specific capacitance (301 F g^{-1} in a three-electrode and 176 F g^{-1} in a couple-electrode system) and exceptional retention stability (92.6% over 10 000 cycles), making it a promising candidate for highly efficient supercapacitors.

2.5. Chemical stability

The chemical and thermal stability of CMPs is a defining characteristic that sets them apart from many other organic materials.¹⁰³ This stability arises from their robust covalent networks, which are formed through strong covalent bonds and the absence of labile functional groups in the polymer backbone. The exceptional stability of CMPs ensures their durability under a diverse array of operating conditions, rendering them highly suitable for supercapacitor systems.



2.5.1. Thermal stability. CMPs exhibit remarkable thermal stability, often withstanding temperatures above 400 °C without significant degradation.^{65,104} This property is critical for ensuring the durability of CMP-based electrodes under high-temperature operating conditions of energy storage devices.¹⁰⁵ The thermal stability of CMPs is attributed to their rigid, three-dimensional covalent networks, which are composed of strong covalent bonds and aromatic building blocks. These networks resist thermal decomposition, even at elevated temperatures, due to the high bond dissociation energies of the covalent linkages.¹⁰⁶ Actually, these thermal features could be enhanced by the stacking energy as well as their planarity.^{20,94} The ability of CMPs to maintain their structural integrity at high temperatures ensures consistent performance in supercapacitors, even under demanding conditions. For example, CMPs have been shown to retain their porosity and surface area after prolonged exposure to high temperatures, rendering them efficient throughout high-power and high-energy applications.¹⁰⁷

2.5.2. Electrochemical stability. The electrochemical stability of CMPs is a key requirement for their use as supercapacitor electrodes. The conjugated framework of CMPs resists electrochemical degradation during repeated charge–discharge cycles, ensuring long-term performance and cycling stability. In addition, the extended π -conjugation in CMPs provides a stable electronic structure that can withstand redox reactions without significant degradation.¹⁰⁸ The electrochemical stability of CMPs enables them to maintain high capacitance and energy density over thousands of charge–discharge cycles. Recently, our group highlights the exceptional performance of a spherical TTh-Ph-PyTE CMP over the corresponding chain polymer as shown in Fig. 8, a CMP integrating redox-active modified Py and

nitrogen-rich triazine, which achieves a remarkable specific capacitance of 1041 F g⁻¹ at 1 A g⁻¹ and outstanding cycling retention with 90% capacitance after 5000 cycles at 10 A g⁻¹, underscoring its promise as an efficient supercapacitor electrode coating.¹⁰⁹ This robust stability is key to the effective application of CMPs in supercapacitors, where long-term reliability is essential.

2.5.3. Resistance to harsh conditions. CMPs are highly resistant to harsh chemical environments, including acidic, basic, and oxidative conditions. This resistance makes them suitable for use in a wide range of electrolytes and operating conditions, further enhancing their versatility in energy storage systems. The robust covalent networks of CMPs resist hydrolysis and degradation in both acidic and basic environments. This property allows CMPs to be used with a variety of electrolytes, including aqueous acids and bases, without compromising their structural integrity.^{43,110,111} CMPs are also stable under oxidative conditions, which is particularly important for supercapacitors operating at high voltages. The conjugated framework resists oxidation, ensuring that the material retains its electrochemical performance even under aggressive operating conditions. Notably, the resistance of CMPs to harsh conditions enables their use in diverse supercapacitor configurations, including aqueous, organic, and ionic liquid electrolytes.¹¹² This versatility allows for the optimization of supercapacitor performance based on specific application requirements, such as high energy density or wide operating voltage windows. On the other hand, although CMPs are generally stable in aqueous environments, prolonged exposure to highly acidic or basic electrolytes may lead to gradual degradation. Furthermore, the synthesis of CMPs with high

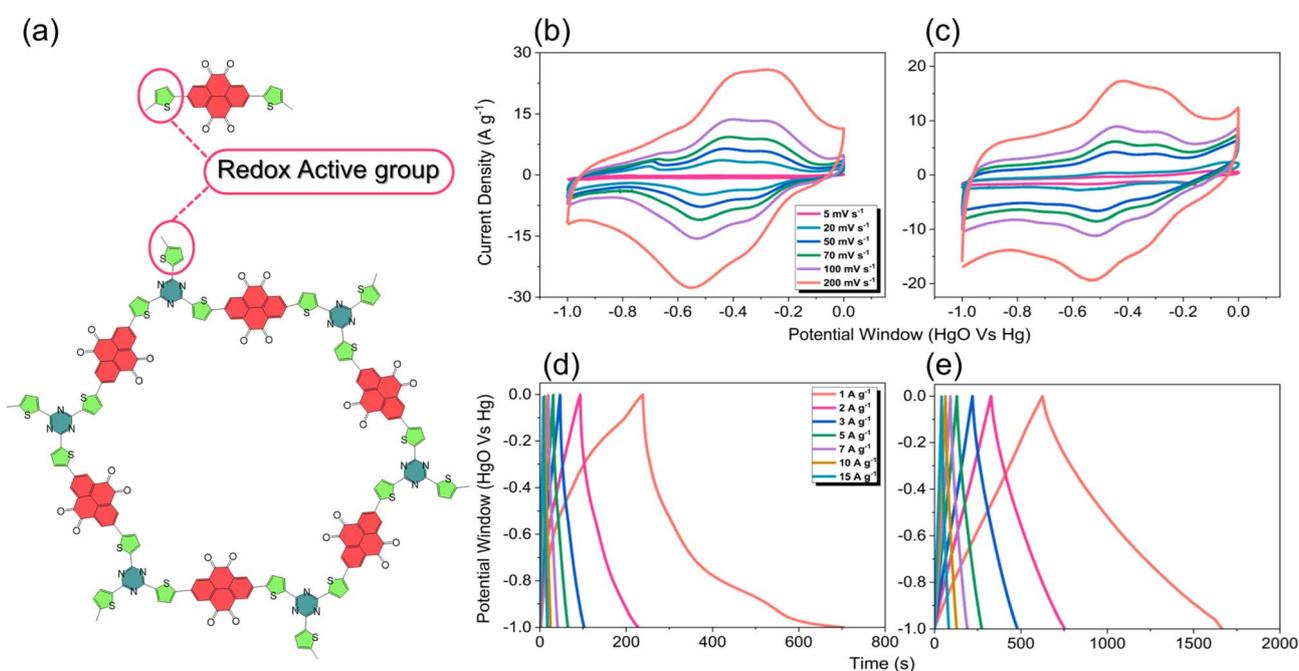


Fig. 8 (a) Chemical structures of the PyTE-Th polymer and TTh-Ph-PyTE CMP, (b and c) cyclic voltammograms and (d and e) galvanostatic charge–discharge profiles of the PyTE-Th polymer (b and d) and TTh-Ph-PyTE CMP (c and e), respectively.¹⁰⁹ Adapted with authorization of Elsevier.



stability often involves complex reactions and expensive catalysts, which can limit their scalability and cost-effectiveness. Developing greener and more scalable synthetic methods is essential for commercialization. In addition, ensuring the prolonged operational stability of CMPs under extreme criteria, such as high voltages, elevated temperatures, and aggressive electrolytes, requires further investigation. Cutting-edge analytical methods and rapid aging tests can contribute to a deeper understanding of the processes leading to material breakdown and inform the design of more stable materials. Therefore, we believe that future research should focus on addressing these defects through innovative material design,

advanced synthesis techniques, and comprehensive stability testing. The continued development of stable CMPs holds great promise for advancing supercapacitor technology and enabling next-generation energy storage solutions.

2.6. Significance in supercapacitor performance

The high capacitance of CMPs is a result of their distinctive blend of large surface area and redox activity, which allows for efficient charge storage through both EDLC and pseudo-capacitance. As we mentioned above, CMPs typically exhibit high surface areas providing abundant active sites for ion adsorption where the electrode and electrolyte interface. This

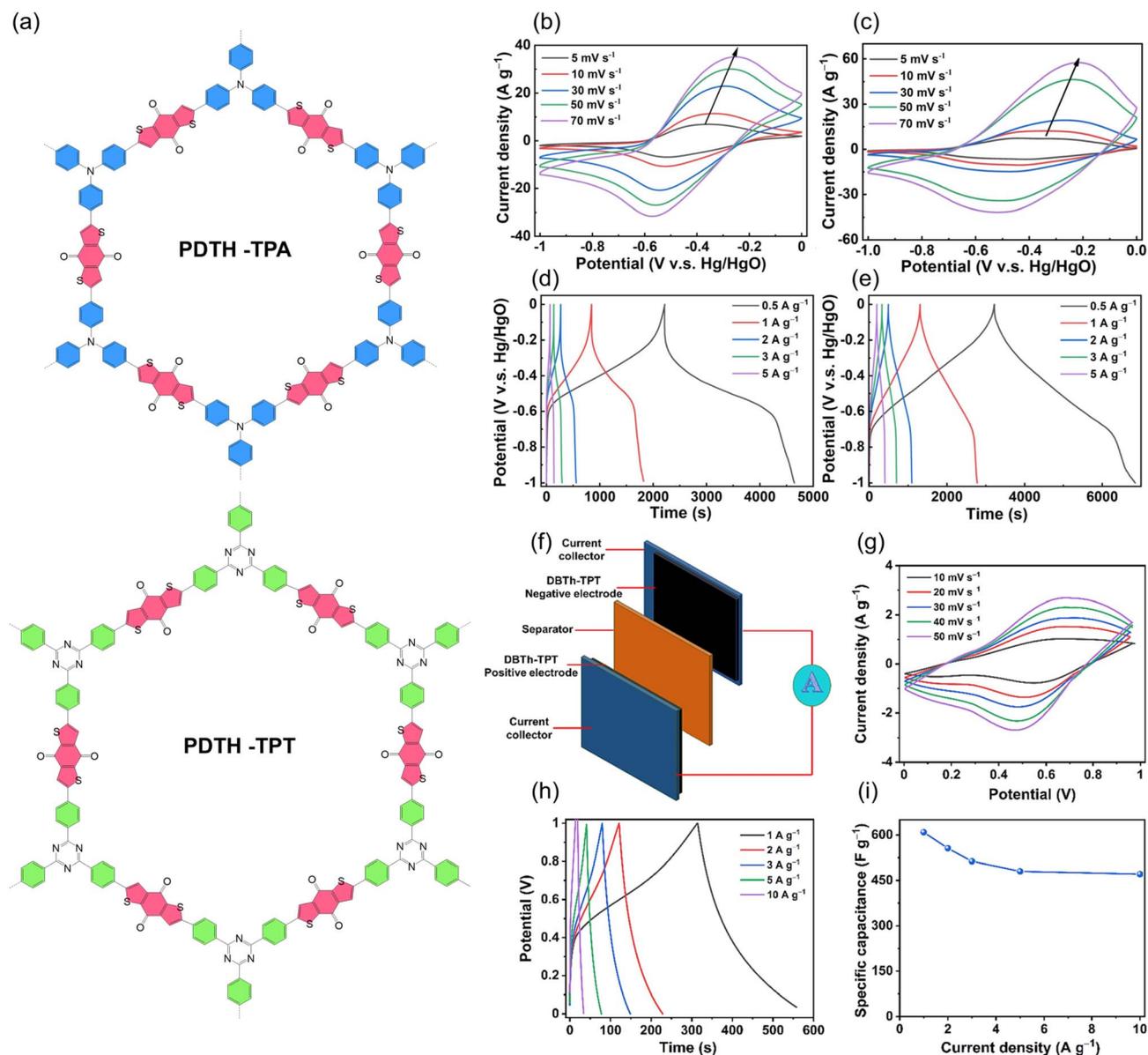


Fig. 9 (a) Molecular structures of DBTh-TPA and DBTh-TPT CMPs; (b) CV curves of the DBTh-TPA CMP, (c) CV curves of the DBTh-TPT CMP recorded at varying scan rates, and (d) GCD profiles of the DBTh-TPA CMP and (e) DBTh-TPT CMP measured at different current densities. (f) Schematic representation of the DBTh-TPT CMP-based supercapacitor device, employing carbon paper as the current collector and filter paper as the separator, (g) CV curves and (h) GCD profiles of the DBTh-TPT CMP-based supercapacitor device at various scan rates and current densities, respectively, and (i) plot of specific capacitance values of the device derived from GCD data across different current densities.³⁶ Adapted with authorization of Elsevier.



high surface area is a critical factor for maximizing EDLC, as it allows for the creation of a large electric double layer, which stores charge electrostatically. On the other hand, the π -conjugated framework of CMPs, often enhanced by the incorporation of heteroatoms or functional groups, enables redox reactions that contribute to pseudocapacitance. Zhan *et al.*³⁶ demonstrated the successful synthesis of oxidation-reduction benzo[1,2-b:4,5-b']dithiophene-4,8-dione-based CMPs (DBTh-CMPs) as shown in Fig. 9, which leverage faradaic processes to achieve an exceptional specific capacitance of 1823 F g^{-1} at 0.5 A g^{-1} using a three-electrode setup, along with remarkable cycling stability (81.75% over 10 000 runs), highlighting their potential as efficient supercapacitor materials with efficient charge transfer and faradaic energy storage capabilities. These faradaic reactions involve the transfer of electrons and ions, providing additional charge storage beyond EDLC.

Therefore, such a combination of EDLC and pseudocapacitance in CMPs results in higher specific capacitances compared to traditional carbon-based materials. Wang *et al.*¹¹³ reported the successful incorporation of CMPs onto multiwall carbon nanotubes (MWNTs), forming MWNT@CMP core-shell composites (Fig. 10) showing a high surface area as well as pore volume. The composites displayed a remarkable capacitance of 248.4 F g^{-1} , significantly outperforming porous carbon. Further carbonization of CMPs onto MWNTs resulted in N- as well as S-doped MWNT@CMP-C composites, further enhancing capacitance due to increased electrochemical sites. The work underscores the importance of integrating EDLC and pseudocapacitance to optimize supercapacitor performance.

The rapid charge/discharge capabilities of CMP-based supercapacitors are enabled by their extended π -conjugation and hierarchical porosity, which facilitate fast ion transport and electron transfer. Also, the delocalized π -electrons in CMPs

create a continuous pathway for efficient electron transport along the polymer backbone. This reduces charge transfer resistance and enables accelerated charge-discharge sequences, which are important for achieving outstanding power density.³² The hierarchical pore structure of CMPs enhances ion transport and electrolyte accessibility. Micropores offer a large surface area for ion adsorption, while mesopores as well as macropores facilitate ion diffusion, reducing resistance, which improves rate capability. Therefore, the combination of extended π -conjugation and hierarchical porosity allows CMP-based supercapacitors to achieve high power densities. This makes them suitable for applications requiring rapid energy delivery, such as electric vehicles and portable electronics.¹⁷ Despite the extended π -conjugation of CMPs, their conductivity is often lower than that of inorganic materials. Developing strategies to further enhance conductivity, such as hybridization with conductive materials or doping, is an ongoing area of research.

3. Mechanisms of energy storage in CMPs

The energy storage mechanisms in CMP-based supercapacitors are primarily governed by two well-known processes, nominated EDLC and pseudocapacitance or even their hybrid. These mechanisms work synergistically to enhance the overall capacitance, energy density, and power characteristics of the device. EDLCs accumulate charge through electrostatic attraction and release of ions on the electrode surface without involving any faradaic redox reactions. Because this process is purely capacitive and non-faradaic, the current response during cyclic voltammetry is proportional to the scan rate and changes smoothly with voltage, resulting in a characteristic rectangular-shaped CV

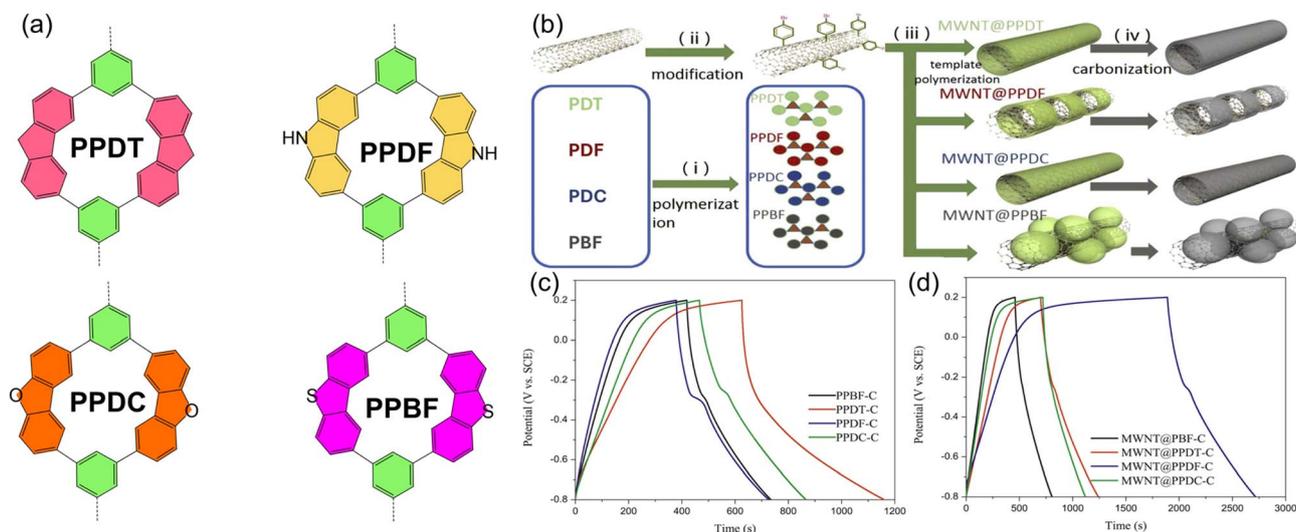


Fig. 10 (a) Molecular structures of PPDT, PPDF, PPDC, and PPBF-based CMPs and (b) synthetic pathway for CMPs, MWNT@CMP composites, and their corresponding porous carbon materials. The procedure includes: (i) palladium-catalyzed coupling under nitrogen at $100 \text{ }^\circ\text{C}$ for 24 h; (ii) reaction with 4-bromoaniline under nitrogen at $80 \text{ }^\circ\text{C}$ for 24 h; (iii) further $\text{Pd}(\text{PPh}_3)_4$ -catalyzed coupling under nitrogen at $100 \text{ }^\circ\text{C}$ for 24 h; and (iv) thermal treatment involving pyrolysis at $450 \text{ }^\circ\text{C}$ for 30 min followed by carbonization at $800 \text{ }^\circ\text{C}$ for 120 min, (c) GCD curves of CMP-derived carbons (CMPs-C) at a current density of 0.5 A g^{-1} , and (d) GCD profile of MWNT@CMP-derived carbons (MWNT@CMP-Cs) recorded at 0.5 A g^{-1} .¹¹³ Adapted with authorization of Elsevier.



curve without distinct peaks as displayed in Scheme 3. In contrast, pseudocapacitive and battery-type materials involve faradaic charge transfer through redox reactions. These redox processes are associated with specific potentials where oxidation and reduction occur, producing well-defined peaks in the CV curves. Pseudocapacitive materials exhibit fast, surface or near-surface redox reactions with broad, symmetrical peaks and minimal voltage separation due to rapid kinetics and minimal diffusion limitations. Battery-type materials, however, involve slower, diffusion-controlled bulk redox reactions often accompanied by phase changes, leading to sharper and more pronounced peaks with larger voltage separations. Thus, the presence or absence of peaks in CV curves reflects the fundamental difference between non-faradaic capacitive behavior in EDLCs and faradaic redox processes in pseudocapacitive and battery-type electrodes (Scheme 3).

3.1. EDLC

The EDLC mechanism is a non-faradaic process that relies on the physical adsorption and desorption of electrolyte ions at the electrode–electrolyte interface. This mechanism is highly dependent on the surface area, pore structure characteristics, and the electrode's conductive properties. CMPs are particularly well-suited for EDLC owing to their efficient high surface area, microporous structure, and chemical durability.¹¹⁴

3.1.1. Ion adsorption at the electrode surface. The EDLC mechanism involves the creation of a Helmholtz double layer at the electrode–electrolyte interface, where electrolyte ions are physically accumulated onto the electrode interface (Scheme 2a). This process is driven by the application of an external voltage, which causes cations and anions to move toward the oppositely charged electrodes. Actually, when a voltage is applied, cations are attracted to the cathode, while anions are drawn to the anode. These ions accumulate at the electrode surface, forming a Helmholtz double layer. The charge segregation occurring at the boundary creates a capacitive effect, storing energy electrostatically. Notably, the high surface area of CMPs offers abundant positions for ion adsorption, leading to a large interfacial area and, consequently, high capacitance. Those CMPs with high surface areas have demonstrated specific capacitances outperforming many traditional carbon-based materials.¹¹⁵

3.1.2. Role of porosity. The porosity of CMPs plays a critical role in optimizing the EDLC mechanism by ensuring efficient ion accessibility and short diffusion pathways. The microporous structure of CMPs, with pore sizes typically less than 2 nm, is particularly effective in accommodating solvated ions and maximizing the interfacial area for charge storage. Furthermore, the narrow pore sizes of CMPs are well-matched to the size of solvated ions in the electrolyte, ensuring efficient ion accessibility. This is critical for maximizing the interfacial area and, consequently, the capacitance. The microporous structure of CMPs provides short diffusion pathways for ions, reducing the resistance to ion transport and enabling quick charging and discharging sequences. This is important for achieving high power density and excellent rate capability.¹¹⁶

3.1.3. Electrolyte compatibility. As we noticed above, the chemical stability of CMPs allows them to operate in a wide range of electrolytes, such as water-based, organic solvent-based, and ionic liquid-based systems. This versatility enhances their applicability in different supercapacitor configurations and operating conditions. CMPs are stable in aqueous electrolytes, such as H₂SO₄ or KOH, which are widely utilized in supercapacitors owing to their ionic conductivity as well as affordability. Moreover, CMPs can also operate in organic electrolytes, which provide a broader voltage window as well as higher energy densities compared to aqueous electrolytes. The chemical stability of CMPs ensures their compatibility with organic solvents, such as acetonitrile or propylene carbonate. On the other hand, ionic liquids are another class of electrolytes compatible with CMPs that offer broad voltage ranges and outstanding energy densities, rendering them suitable for efficient supercapacitor designs.^{117,118}

3.2. Pseudocapacitance

Pseudocapacitance is a faradaic process that involves reversible redox interactions near the electrode interface (Fig. 5). Unlike EDLC, pseudocapacitance stores charge *via* chemical processes, resulting in outstanding energy densities. CMPs exhibit pseudocapacitive behaviour due to their conjugated π -systems and the presence of redox-active functional groups or heteroatoms.¹¹⁹

3.2.1. Redox-active functional groups. The incorporation of heteroatoms within the CMP framework introduces redox-active sites that participate in faradaic processes. These heteroatoms can undergo reversible redox transitions between different oxidation states, contributing to additional charge storage. Nitrogen doping is one of the most effective strategies for enhancing pseudocapacitance in CMPs. Further, such nitrogen atoms can exist in various configurations, including pyridinic nitrogen, pyrrolic nitrogen, and quaternary nitrogen, each of which contributes to redox activity.¹²⁰ Zhang *et al.* explored the design of p/n-dopable conducting redox polymers utilizing redox-active groups incorporated within the polymer backbone or attached as side chain groups, demonstrating enhanced cycling stability and energy storage capabilities for supercapacitors.¹²¹ The study highlighted that nitrogen doping in these polymers can significantly increase pseudocapacitance by facilitating redox reactions and improving charge storage, while also addressing the charge-trapping effect during n-doping, which is a key factor in performance degradation. Also, pyrrolic-N enhances electron donation and improves charge transfer kinetics.¹²² Quaternary-N increases the overall conductivity of the material.¹²³ Oxygen- and sulfur-containing functional groups, such as quinones, hydroquinones, and thiols, also exhibit redox activity. These groups undergo reversible redox reactions, providing additional charge storage capacity.

3.2.2. Conjugated π -system. The extended π -conjugation in CMPs plays a critical role in enhancing pseudocapacitive performance by facilitating efficient charge transfer and electron delocalization. The overlap of π -orbitals in the conjugated



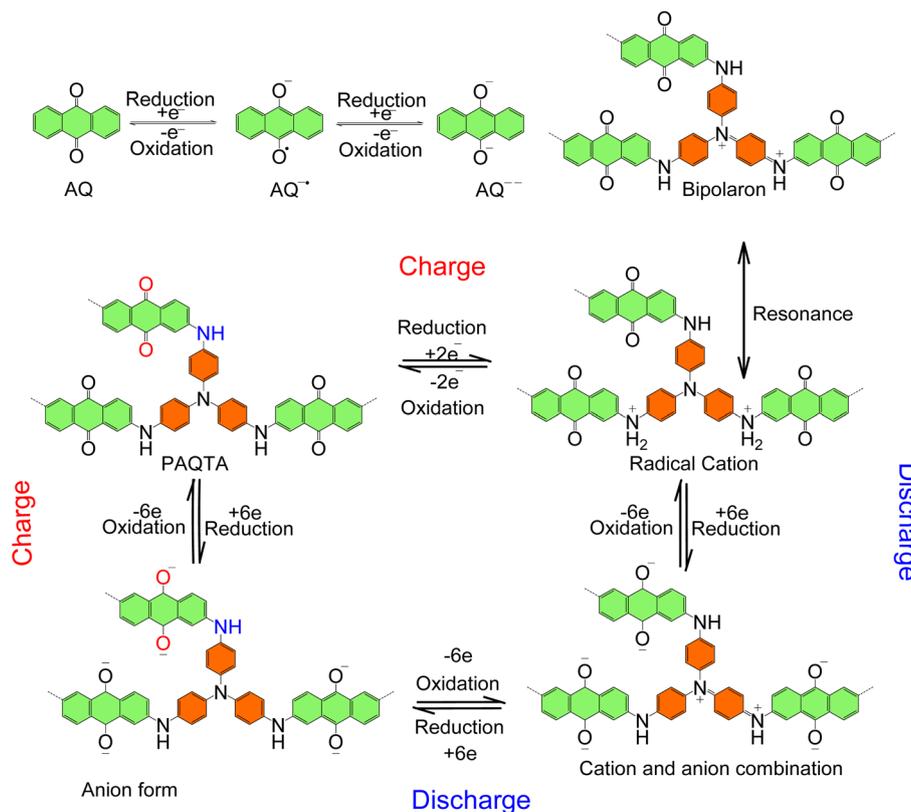


Fig. 11 Illustration of the charge–discharge mechanism and the associated redox reactions proposed for PAQTA.¹¹⁷

framework creates a delocalized electron system, which reduces the energy barrier for electron movement. This delocalization enables rapid charge transfer during redox reactions, improving reaction kinetics and reducing charge transfer resistance.¹²⁴ The combination of redox-active functional groups and the conjugated π -system creates a synergistic effect, enhancing both the energy density and power density output of CMP-based supercapacitors. For example, nitrogen-doped CMPs with extended π -conjugation have demonstrated specific capacitances with a significant contribution from pseudocapacitive processes.⁹⁴

3.2.3. Surface functionalization. The introduction of redox-active functional groups through surface functionalization further enhances the pseudocapacitive behavior of CMPs. These groups undergo reversible redox reactions, providing additional charge storage capacity. Notably, quinone groups can reversibly transition between the quinone and hydroquinone states, contributing to pseudocapacitance. This redox activity is highly reversible, ensuring long-term cycling stability. The study of PAQTA as an electrochemical energy storage material highlights its exceptional pseudocapacitive performance, which is primarily governed by a synergetic redox mechanism involving anthraquinone (AQ) and triphenylamine (TPA) moieties.¹¹⁷ The redox reaction of AQ follows a two-electron reduction to form a dianion, while the PAQTA polymer undergoes oxidation to form polaron and bipolaron states as displayed in Fig. 11, enhancing charge storage capacity. The integration of these redox-active units results in an exceptional theoretical

capacitance of 1440 F g^{-1} , though only $\sim 40\%$ of the repeating units actively participate in the redox process. The asymmetric supercapacitor (ASC) coin incorporating PAQTA with commercial activated carbon (AC) demonstrates a stable voltage range of 0–1.6 V, with a specific capacitance of 168 F g^{-1} at 1.0 A g^{-1} and remarkable rate capability (106 F g^{-1} at 10 A g^{-1}). The redox mechanism is further validated by FT-IR characterization, showing reversible transformations between benzenoid and quinoid structures with high coulombic efficiency (90.5–93.5%).

The DBTh-based CMPs in this study exhibit exceptional pseudocapacitance, driven by a synergistic redox mechanism involving DBTh, TPA, and TPT moieties, as shown in Fig. 12. Their charge storage is dominated by reversible two-electron redox reactions, as confirmed by CV, *ex situ* FTIR, and EIS analyses.³⁶

The redox mechanism of CC-DAQ-CMP is driven by complementary interaction through redox-active DAQ units and nitrogen-rich conductive CC components. The charge storage process incorporates the reversible two-electron reduction of AQ to AQ^{2-} anions and the redox mobility between aryl and quinone structures in the cyanuric chloride (CC) monomers. During discharge, AQ and quinone groups are reduced, while charge transfer and oxidation of AQ^{2-} and nitrogen-rich cyanuric chloride units occur during charging (Fig. 13a). This dual redox process enhances charge transfer efficiency and energy storage.¹²⁵ In addition, our recent study demonstrates the pivotal role of pseudocapacitance in enhancing the



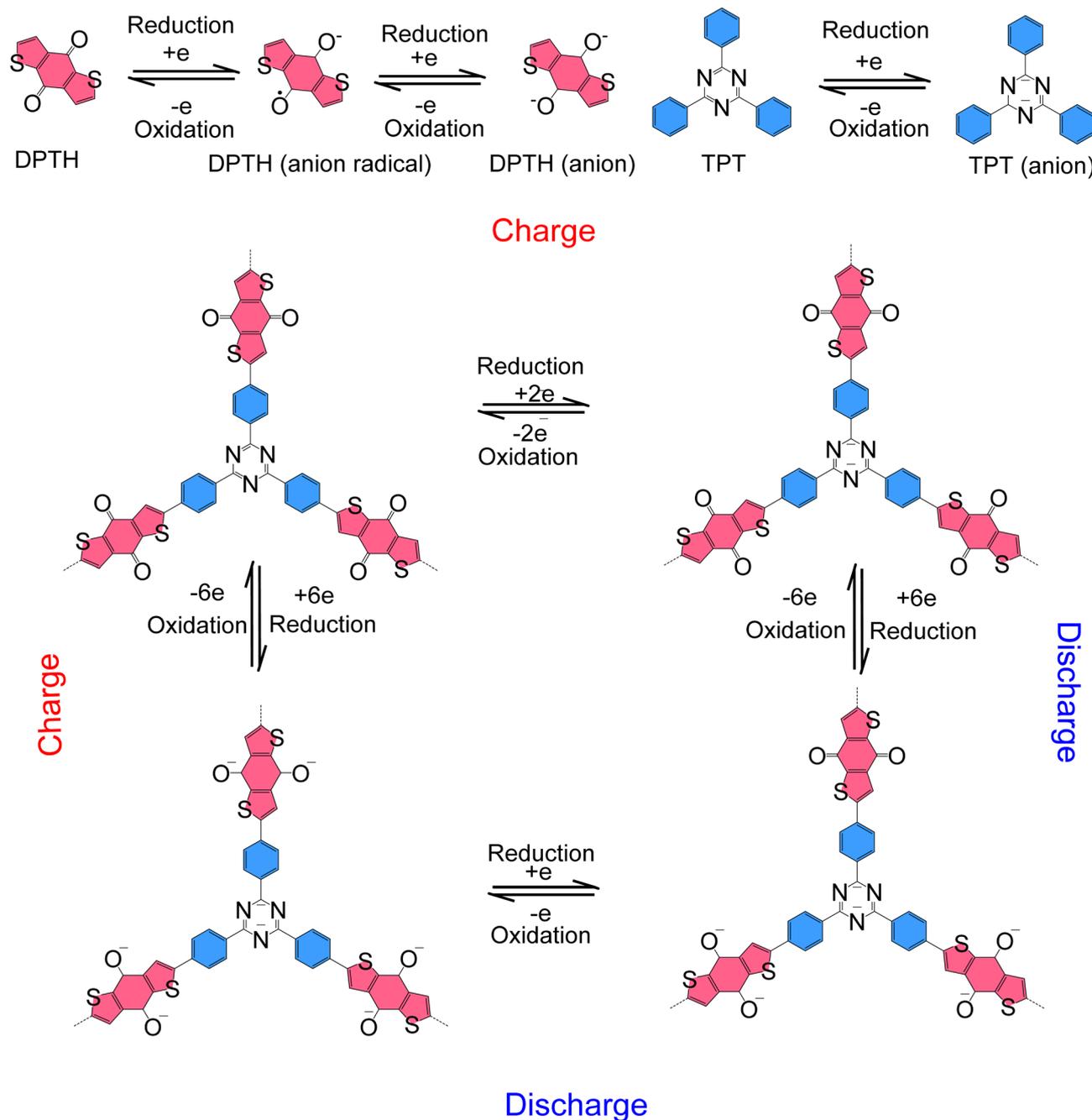


Fig. 12 Proposed redox mechanisms for DBTh, TPT, and the DBTh-TPT CMP.³⁶

electrochemical performance of redox-active phenazine-based CMPs, where the superior planarity and imine-rich structure of TPT-QP CMP enable effective accumulation of charge *via* a faradaic mechanism involving the reversible reduction of QP and TPT to QP⁴⁻ and TPT²⁻ anions (Fig. 13b).²⁰

Importantly, thiol groups can be oxidized and reduced reversibly, forming and breaking disulfide bonds, providing additional redox activity. This process is particularly effective in sulfur-doped CMPs, which exhibit high pseudocapacitance and energy density.^{126,127} Therefore, we believe that functional groups such as amines, carboxylates, and imines can also be

utilized to enhance pseudocapacitance. These groups offer extra sites for redox reactions and improve the total electrochemical efficiency of CMPs. The reversible nature of the redox reactions in pseudocapacitance ensures long-term cycling stability, with many CMPs retaining more than 90% of their initial capacitance over 5000 runs. Otherwise, the faradaic nature of pseudocapacitance can lead to slower reaction kinetics compared to EDLC, particularly at high charge-discharge rates. This can limit the power density of pseudocapacitive materials. Further, while pseudocapacitance is generally reversible, the repeated redox reactions can lead to gradual degradation of the



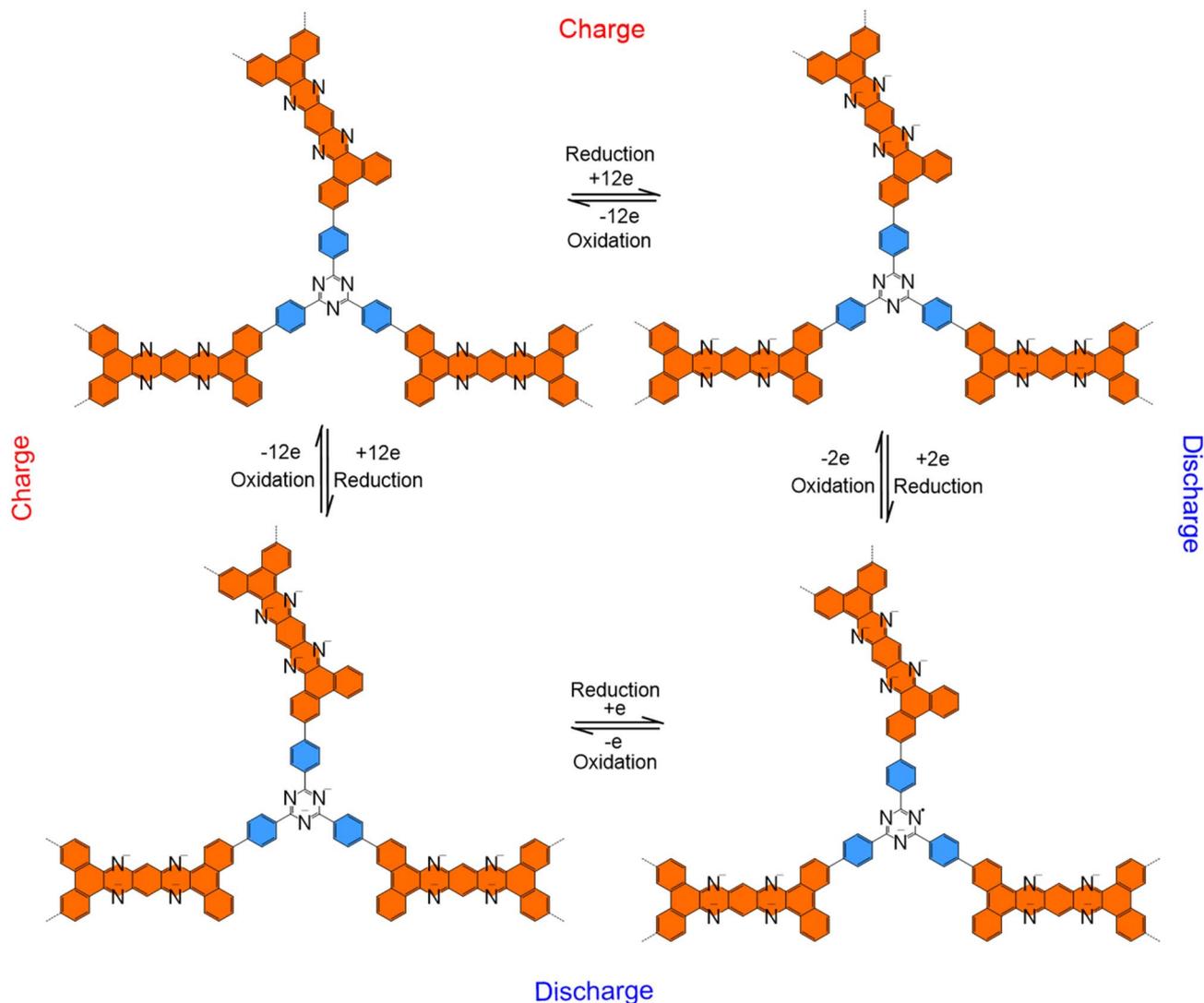


Fig. 13 (a) Schematic representation of the redox electron transfer mechanism during the charge and discharge processes occurring within nitrogen-rich cyanuric chloride units.¹²⁵ (b) Proposed redox pathways for QP, TPT, and the resulting TPT-QP CMP.²⁰

electrode material over time. Ensuring long-term cycling stability is a key challenge.

3.3. Synergistic effects of EDLC and pseudocapacitance

In CMP-based supercapacitors, the EDLC and pseudocapacitance mechanisms often operate simultaneously, leading

to a synergistic enhancement of energy storage performance. The high surface area and porosity of CMPs facilitate efficient ion adsorption (EDLC), while the redox-active sites and conjugated π -systems enable faradaic reactions (pseudocapacitance). The coexistence of EDLC and pseudocapacitance allows CMPs to achieve higher specific capacitance compared to materials



relying solely on one mechanism. In addition, the synergistic effects enable CMP-based supercapacitors to deliver both high energy density (from pseudocapacitance) and high power density (from EDLC). Furthermore, the robust covalent framework of CMPs ensures structural integrity through successive charging and discharging processes, mitigating the degradation often associated with pseudocapacitive materials.

3.4. Factors influencing energy storage mechanisms

The energy storage performance of CMPs is influenced by several parameters including (1) pore size and distribution where the optimal pore sizes (micropores for ion adsorption and mesopores for ion transport) are critical for maximizing both EDLC and pseudocapacitance, (2) heteroatom doping where the type and concentration of heteroatoms determine the redox activity and pseudocapacitive contribution, (3) selecting between aqueous, organic, and ionic liquid electrolytes affecting the ion mobility, operating voltage, and overall capacitance, and (4) the design of the electrode (*e.g.*, thin films, composites, or 3D structures), which influences ion diffusion, conductivity, and mechanical stability.

4. Recent advances in CMPs for supercapacitor applications

Over the past few years, significant progress has been made in optimizing the efficiency of supercapacitors built with CMPs, driven by innovative synthetic strategies, advanced material design, and novel device engineering approaches. This section surveys recent studies, highlighting key advancements in key indicators like capacitance, energy output, power capability, and cycling stability, while comparing CMPs to traditional materials like activated carbon, graphene, and metal oxides.

4.1. Enhanced capacitance through structural design

Recent research has shown that the capacitance of CMP-based supercapacitors can be significantly enhanced through precise control over their structural and chemical properties. For example, researchers have developed CMPs with high surface areas achieved through the use of rigid, planar monomers and efficient coupling reactions. Our earlier study demonstrates that the high surface area of TPA-based CMPs, particularly TPA-Py CMP ($1470 \text{ m}^2 \text{ g}^{-1}$), has a significant impact on their superior capacitive performance, highlighting the direct correlation between enhanced porosity, structural planarity, and improved supercapacitor capacitance.⁷⁰ In addition, the introduction of hierarchical porosity (micro-, meso-, and macropores) in CMPs is proven to facilitate ion transport and accessibility. Zhang *et al.*¹²⁸ highlighted that the hierarchical porosity of porphyrin-based CMP gels integrated with a graphene hydrogel (GH) significantly enhances mass transfer and ion diffusion, resulting in a high specific capacitance of 208 F g^{-1} and excellent cycling stability with 92.6% capacitance retention after 10 000 cycles, making GH-CMP a promising electrode material for supercapacitors. Once again, the incorporation of redox-active functional groups or heteroatoms into the CMP framework has enhanced pseudocapacitance. Within this regard, Jang *et al.*¹²⁹ highlighted the critical role of functionalizing CMPs with redox-active anthraquinonylamine units and their integration with activated graphene to enhance electrochemical behaviour as demonstrated in Fig. 14. Through controlled acid treatment of graphene, the optimized CMAP@A4G hybrid exhibited a large surface area of $498 \text{ m}^2 \text{ g}^{-1}$, improved electrical conductivity, and superior physicochemical stability. The incorporation of anthraquinonylamine and TPA into the CMP network facilitated fast electron transfer and more redox-active centres, leading to an exceptional specific capacitance of 751 F g^{-1} at 1.0 A g^{-1} and outstanding cycling stability, retaining 97% of capacitance after 20 000 cycles at 10 A g^{-1} . Furthermore, asymmetric supercapacitors incorporating CMAP@A4G showed

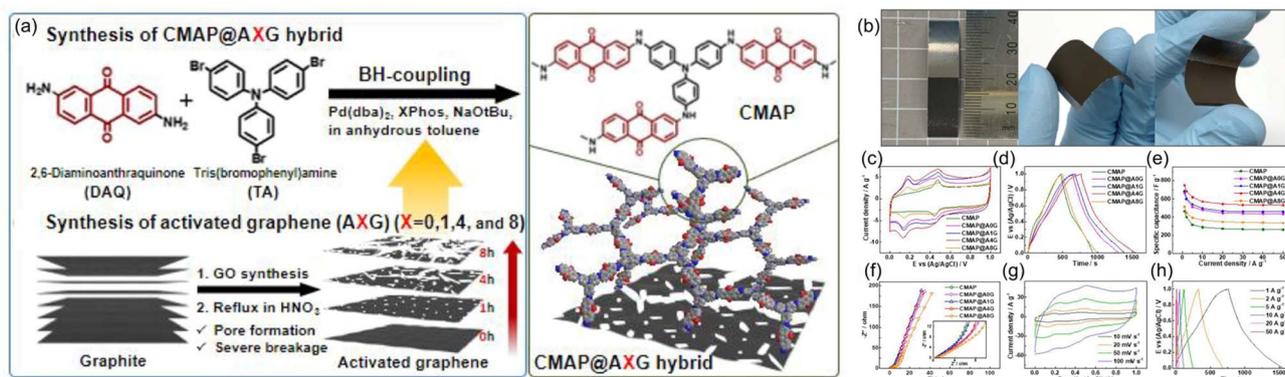


Fig. 14 (a) Schematic depiction of the synthesis of AXG and CMAP@AXG hybrid materials, where 'X' represents the acid treatment duration (0, 1, 4, and 8 hours, respectively), (b) photographic images showing the flexibility of CMAP@A4G electrodes, (c–h) electrochemical characterization of CMAP, CMAP@A0G, CMAP@A1G, CMAP@A4G, and CMAP@A8G in a three-electrode setup using 1.0 M H₂SO₄ electrolyte: (c) CV curves at 10 mV s^{-1} , (d) GCD profiles at 1 A g^{-1} , (e) specific capacitance values across scan rates from 1 to 50 A g^{-1} , (f) electrochemical impedance spectra (Nyquist plots) over the frequency range of 100 kHz to 10 mHz, with the inset highlighting the high-frequency region, (g) CV curves of CMAP@A4G at varying scan rates, and (h) GCD curves of CMAP@A4G at different current densities.¹²⁹ Adapted with authorization of Elsevier.



a high energy density of 76.6 Wh kg^{-1} , a remarkable power density of $27\,634 \text{ W kg}^{-1}$, and a capacitance retention of 88% after 20 000 cycles at 10 A g^{-1} within a voltage window of 0–1.5 V. These findings underscore the importance of CMP functionalization in enhancing redox activity, conductivity, and electrochemical durability, offering an effective strategy for designing next-generation efficient supercapacitor materials.

4.2. Improved energy density and power density

4.2.1 Hybrid CMP-carbon composites. The integration of CMPs with conductive carbon-based materials such as graphene and carbon nanotubes has resulted in composites with enhanced conductivity and mechanical strength. Our group *et al.*¹¹⁰ employed a conventional Sonogashira coupling reaction to synthesize two pyrene-linked CMPs, designated as PyT-PTCDA and PyT-PHTD CMPs. These were prepared by reacting 1,3,6,8-tetraethynylpyrene (Py-T) with 1,7-dibromo-3,4,9,10-perylenetetracarboxylic dianhydride (PTCDA-Br₂) and 3,6-dibromophenanthrene-9,10-dione (PHTD-Br₂), respectively. Comprehensive characterization using spectroscopy and microscopy confirmed the successful formation of these CMPs. Notably, the PyT-PTCDA CMP exhibited high thermal resistance, decomposing at 351 °C and leaving a char yield of 61 wt%. To enhance their electrical conductivity and electrochemical properties, we integrated these PyT-CMPs with highly conductive single-walled carbon nanotubes (SWCNTs) *via* π - π stacking interfaces of the PyT units and SWCNTs. Electrochemical assessments confirmed that the PyT-PTCDA CMP/SWCNT nanocomposite delivered a remarkable specific capacitance of 376 F g^{-1} at 0.5 A g^{-1} in a three-electrode system while retaining 98% of its capacitance after 5000 charge-discharge runs. Additionally, the composite displayed an energy density of 52 Wh kg^{-1} in a three-electrode setup and 17 Wh kg^{-1} in a symmetric device, with a corresponding capacitance of 119 F g^{-1} . This work underscores the critical role of integrating CMPs with conductive carbon materials, such as SWCNTs, in significantly improving charge transport, electrochemical stability,

and overall supercapacitor performance. The synergy between the porous CMP networks and highly conductive SWCNTs provides a promising strategy for upcoming generations of energy storage devices.

4.2.2 CMP-metal oxide hybrids. Combining CMPs with redox-active metal oxides has further boosted energy density. COFs have emerged as a promising family of crystalline CMPs due to their well-ordered channels, high surface areas, and tuneable active sites, rendering them efficient for energy storage applications. However, their practical implementation is hindered by inherent low conductivity and poor processability. To overcome these limitations, a cation-driven self-assembly strategy was employed to fabricate a flexible composite film electrode (CMF) by integrating carbonyl-functionalized COFs with MXene nanosheets. The electrostatic attraction of negatively charged MXenes with protonated DAAQ-COFs facilitated uniform intercalation as displayed in Fig. 15, preventing the undesirable restacking of MXene layers while ensuring intimate contact between the two components.¹³⁰ The synergistic combination of the ordered microporosity of COFs and the high conductivity of MXenes significantly enhanced charge transport, ion migration, and overall electrochemical performance. The resulting CMF electrode exhibited outstanding capacitance (390 F g^{-1} at 0.5 A g^{-1}), superior kinetic energy storage characteristics (up to 96.7% capacitive contribution at 50 mV s^{-1}), and remarkable mechanical stability (withstanding 100 bending cycles). When assembled into an all-solid-state asymmetric supercapacitor (CMFs//CCMP ASC), the device demonstrated a high energy density of 27.5 Wh kg^{-1} at a power density of 350 W kg^{-1} , an increased power density of 7000 W kg^{-1} at an energy density of 19.7 Wh kg^{-1} , and exceptional cycling stability, retaining 88.9% of its capacitance over 20 000 charging/discharging cycles.

4.2.3 Asymmetric supercapacitors. The development of asymmetric supercapacitors, where CMPs serve as the cathode, while carbon-based materials are used as the anode, has enabled higher operating voltages and energy densities.

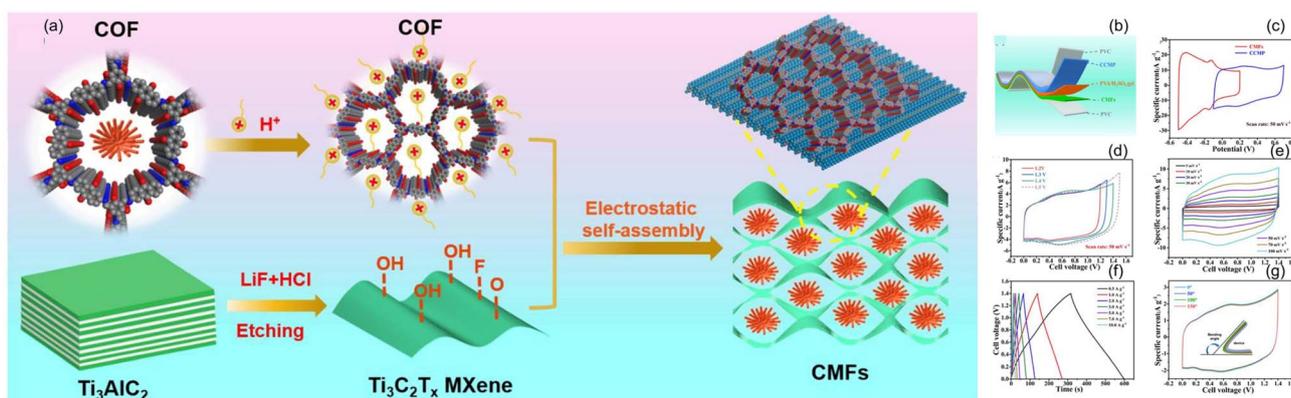


Fig. 15 (a) Schematic illustration of the fabrication process for CMFs. Electrochemical evaluation of the CMFs/CCMP asymmetric supercapacitor (ASC) device, (b) conceptual diagram of the ASC configuration, (c) CV profiles of CMF and CCMP electrodes recorded at 50 mV s^{-1} in a three-electrode setup, (d) CV curves of the ASC measured at 50 mV s^{-1} under various voltage windows, (e) CV curves of the ASC at different scan rates, (f) GCD profiles at multiple current densities, and (g) CV curves of the ASC tested at 10 mV s^{-1} under various bending angles to evaluate flexibility.¹³⁰ Adapted with authorization of Elsevier.



4.3. Enhanced cycling stability and durability

The long-term stability of CMP-based supercapacitors has been a focus of recent research, with significant improvements achieved through advanced material design and device engineering.

4.3.1 Robust covalent frameworks. The inherent chemical stability of CMPs ensures structural integrity during repeated charge–discharge cycles. Ambrose *et al.*¹³¹ reported the synthesis of two distinct viologen-based covalent organic polymers (COPs) with tailored morphologies—COP-1 featuring a hollow sphere structure and COP-2 exhibiting a hollow tubular architecture. The extended π -conjugation and robust structural stability of these COPs effectively address common challenges in polymer-based electrodes, including inhomogeneous aggregation, structural degradation over multiple charge–discharge cycles, and poor interconnectivity during electrode assembly. Among the synthesized materials, COP-2 demonstrated outstanding electrochemical performance when tested as the working electrode in a three-electrode setup supercapacitor design with 1 M H₂SO₄ as the electrolyte as displayed in Fig. 16. It exhibited an impressive specific capacitance of 604 F g⁻¹ at 2 A g⁻¹ and retained 100% of its capacitance even after 50 000 charge–discharge cycles, showcasing remarkable long-term durability. In full-cell studies, COP-2 further delivered a specific capacitance of 404 F g⁻¹ at 0.5 A g⁻¹, underscoring its superior energy storage capability.

4.3.2 Flexible and wearable systems. The development of wearable CMP-based supercapacitors has demonstrated excellent mechanical durability. Teng *et al.*⁴³ reported that the advancement of efficient flexible solid-state supercapacitors (FSSCs) is crucial for the rapid growth of wearable electronic designs. In this study, the Teng group presented an innovative porous layered film electrode made of reduced graphene oxide (rGO) and CMPs to achieve FSSCs with superior specific capacitance and excellent rate capability. This material is fabricated through a simple vacuum filtration method followed by reduction as shown in Fig. 17.

The unique porous sandwich-like architecture has an essential function in increasing the exposure of active sites, facilitating efficient ion mobility, and enhancing electron transport between rGO and CMPs. To address the inherent poor processing of CMPs, a third subunit containing functional hydroxyl was incorporated into the synthesis of a redox-active imine *N*-containing CMP (PTPAH) *via* a one-step cross-coupling reaction. The resulting PTPAH@rGO composite film electrode exhibited a remarkable specific gravimetric capacitance of 545 F g⁻¹ at 1 A g⁻¹, maintaining 450 F g⁻¹ even at a high current density of 10 A g⁻¹. Furthermore, FSSCs assembled with PTPAH@rGO and a PVA/H₂SO₄ gel electrolyte delivered outstanding electrochemical performance, achieving a capacitance of 220 F g⁻¹ and a high-power density of 3345 W kg⁻¹.

4.3.3 Solid-state supercapacitors. The use of gel or solid electrolytes in CMP-based supercapacitors has improved safety as well as stability. Zhang *et al.*¹³² presented a nanoporous 3D conjugated polymer network strategy for developing high-performance, cost-effective flexible electrochromic supercapacitors (FESs). The Zhang team synthesized a conjugated tripod-structured precursor from TPA and 3,4-ethylene-dioxythiophene (EDOT), followed by electro-polymerization to form the resulting polymer (PTEPA). The incorporation of EDOT enhances charge transport and stability, while the nanoporous structure facilitates ion diffusion, improving electrochemical activity. The PTEPA electrode shows excellent electrochemical stability (91.1% retention after 6000 cycles), a high specific capacitance of 137 F g⁻¹ at 1 A g⁻¹, and reliable electrochromic properties with multicolor transitions and high coloring efficiency (136 cm² C⁻¹ at 450 nm). A flexible electrochromic supercapacitor device was successfully fabricated, demonstrating stable energy storage and durability under mechanical bending. This work highlights the potential of 3D conjugated polymer networks for advancing multifunctional energy storage materials.

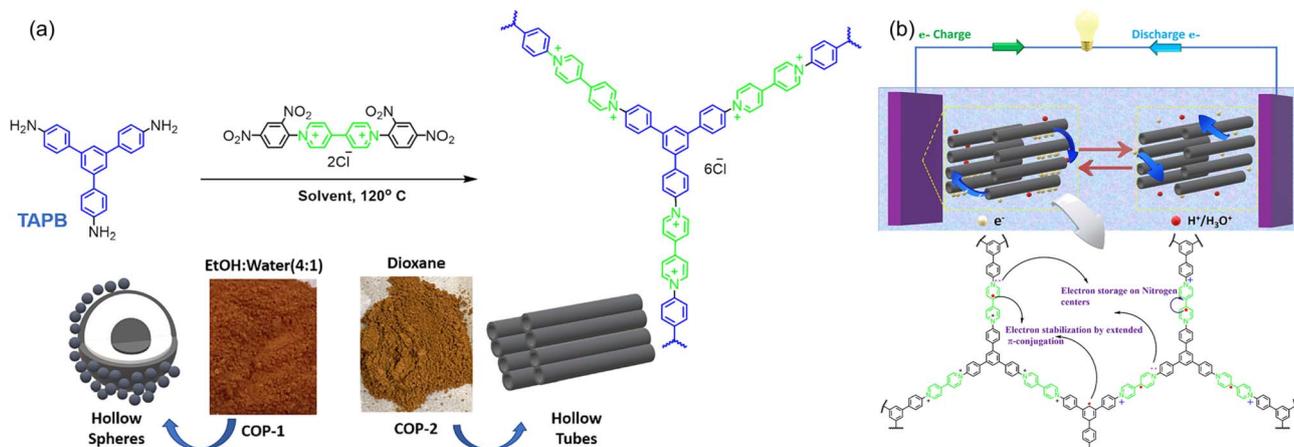


Fig. 16 (a) Schematic representation of the solvothermal synthesis of viologen-based conjugated organic polymers (COPs) through the Zincke reaction and (b) proposed charge transfer and storage mechanism occurring in COP-2 during the electrochemical charging and discharging processes.¹³¹ Adapted with authorization of Elsevier.



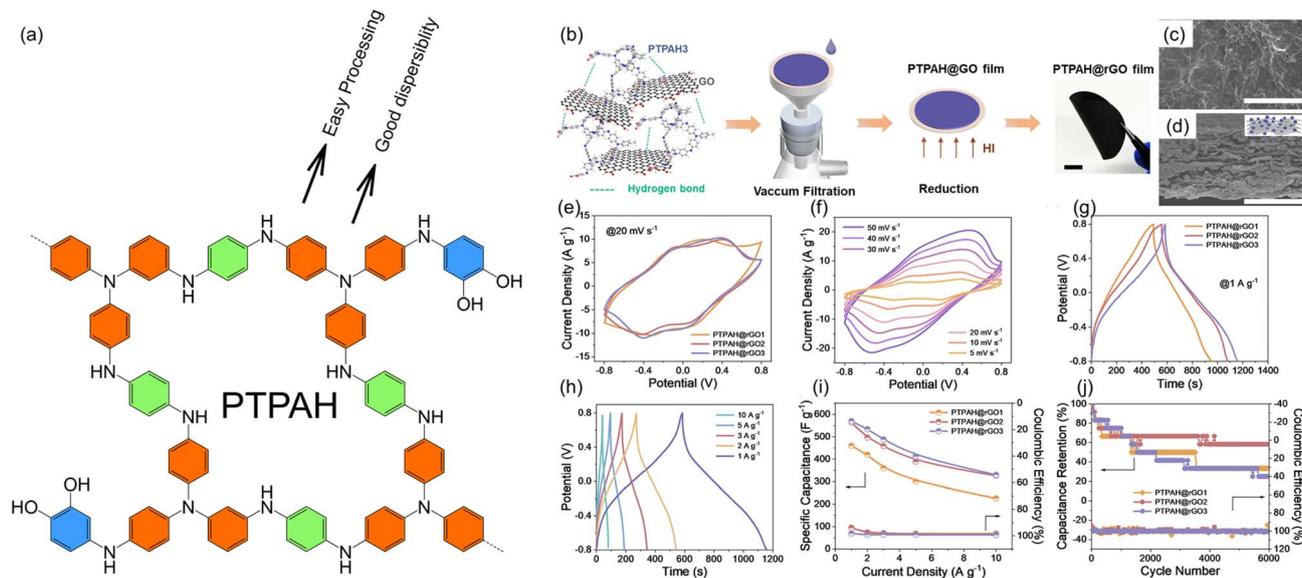


Fig. 17 (a) Illustration highlighting the excellent dispersibility and ease of processing of PTPAH, (b) stepwise procedure for fabricating PTPAH@rGO film composites, with a photo showing the flexibility of the folded film, and (c) top-view and (d) cross-sectional SEM images of the PTPAH@rGO films. Electrochemical performance of PTPAH@rGOs evaluated in a two-electrode configuration: (e) CV curves of three PTPAH@rGO samples recorded at 20 mV s^{-1} , (f) CV profiles of PTPAH@rGO3 at various scan rates, (g) GCD curves of the three PTPAH@rGO samples at 1 A g^{-1} , and (h) GCD curves of PTPAH@rGO3 at different current densities. (i) Comparison of specific capacitance and coulombic efficiency, and (j) cycling retention and coulombic efficiency of PTPAH@rGO composites.⁴³ Adapted with authorization of Royal Society of Chemistry.

4.4. Comparison with traditional materials

CMPs have shown significant advantages over traditional supercapacitor materials in terms of performance metrics.

4.4.1. Activated carbon. While activated carbon is widely used because of its large surface area and low cost, it primarily relies on EDLC, limiting its energy density. CMPs, with their combined EDLC and pseudocapacitance, offer efficient specific capacitance and energy density.¹³³

4.4.2 Graphene. Graphene-based supercapacitors show high conductivity and power density but often suffer from restacking issues, reducing their effective surface area. CMPs, with their rigid and porous structures, avoid restacking and provide more accessible active sites.¹³⁴

4.4.3. Metal oxides. Metal oxides (*e.g.*, RuO_2 and MnO_2) offer high pseudocapacitance but are often limited by poor conductivity and cycling stability. CMP-metal oxide hybrids combine the strengths of both materials to deliver enhanced energy density and stability.¹³⁵

4.5. Emerging trends and future directions

Recent advancements in CMP-based supercapacitors have opened up new avenues for research and development including (1) multifunctional CMPs, where the design of porous polymers with additional functionalities, such as photocatalytic activity or self-healing properties, is an emerging trend.¹³⁶ These multifunctional CMPs could enable integrated energy storage and conversion systems. (2) Designing synthesis approaches that are both economical and suitable for large-scale production for CMPs is critical for their commercialization and we believe that this is a trade-off point. Recent efforts have focused

on green chemistry approaches and template-assisted synthesis to reduce costs and environmental impact. (3) The use of advanced analytical methods including *in situ* spectroscopy and computational modelling provides a better understanding of how charge is stored and guides the design of optimized CMPs.^{21,36} Recent advances in CMP-based supercapacitors have demonstrated significant improvements in performance metrics such as capacitance, energy density, power density, and cycling stability. These advancements highlight the potential of CMPs to outperform traditional materials and address the limitations of current supercapacitor technologies. Through leveraging innovative synthetic strategies, advanced material design, and novel device engineering approaches, researchers are laying the foundation for efficient, durable, and versatile next-generation energy storage technologies. The continuous development of CMPs holds great promise for advancing supercapacitor technology and enabling various uses, including mobile electronics and power grid storage.

5. Hybrid and composite CMP systems

Hybridizing CMPs with complementary active components to form hybrid and composite systems has emerged as a powerful strategy to facilitate the electrochemical behaviour of supercapacitors. By combining the unique properties of CMPs with the complementary characteristics of other materials (*e.g.*, conductivity, mechanical strength, and additional redox activity), these hybrid systems address key challenges such as



low conductivity, insufficient mechanical strength, and insufficient energy density.

5.1 CMP-carbon composites

Graphene, CNTs, and activated carbon are extensively utilized in supercapacitors owing to their superior conductivity, mechanical strength, and chemical stability. Combining CMPs with carbon materials creates composites that leverage the strengths of both components.¹³⁷

5.2 CMP-graphene composites

Graphene, with its exceptional electrical conductivity as well as large surface area, is a good partner for CMPs. In CMP-graphene composites, the CMPs provide redox-active sites and microporosity, while graphene enhances conductivity and mechanical stability. The use of graphene-CMP nanocomposites in this study is driven by their complementary properties. Fig. 18 shows the redox-active Fc-CMPs, which provide abundant faradaic sites and high porosity to enhance charge storage, while monolayer rGO significantly improves electrical conductivity and structural stability. This synergistic incorporation results in exceptional electrochemical activity, showing a capacitance of 470 F g^{-1} (933 mF cm^{-2}) in a three-electrode setup and 231 F g^{-1} (238 mF cm^{-2}) in a couple-electrode device, alongside superior cycling stability (95% retention after 8000 cycles). The use of microporous polymers with redox functionality as well as conductive graphene thus presents a promising strategy for advancing high-performance energy storage devices.⁸⁹

The integration of graphene with CMPs in G-CMP sandwiches significantly enhances capacitance by providing high surface areas, improved conductivity, and hierarchical porosity,

demonstrating a prospecting strategy for developing high-activity supercapacitor materials as displayed in Fig. 19.¹³⁸ The integration of porphyrin-based CMPs with a graphene hydrogel (GH) creates a synergistic composite (GH-CMP) that enhances conductivity, increases surface area, and introduces hierarchical porosity, significantly improving ion diffusion, charge storage, and capacitance.¹²⁸

5.3 CMP-carbon nanotube (CNT) composites

CNTs offer high conductivity and a fibrous structure that facilitates ion transport. CMP-CNT composites combine the microporosity of CMPs with the mesoporous network of CNTs, resulting in enhanced ion accessibility and charge transfer. These composites are particularly effective in high-rate applications owing to their facilitated conductivity and mechanical flexibility. In earlier study, CMPs were synthesized *via* the Sonogashira-Hagihara cross-coupling using a TBN monomer with Py, TPE, and carbazole (Car) units, and their structural, thermal, and electrochemical properties were investigated. Among them, TBN-TPE-CMP exhibited the highest surface area ($1150 \text{ m}^2 \text{ g}^{-1}$) and remarkable thermal stability ($T_{d10} = 505 \text{ }^\circ\text{C}$, char yield = 68 wt%). To enhance the conductivity of these CMPs, they were blended with highly conductive SWCNTs. The derived TBN-Py-CMP/SWCNT nanocomposite demonstrated superior electrochemical performance, achieving a high capacitance of 430 F g^{-1} at 0.5 A g^{-1} and an outstanding cycling retention of 99.18% over 2000 cycles. The integration of SWCNTs with CMPs enhances charge transport and electrochemical activity, making CNT-CMP nanocomposites prospective materials for future energy storage applications.¹³⁹ The combination of CMPs and superior conductive SWCNTs *via* π - π stacking interactions significantly enhances electrical

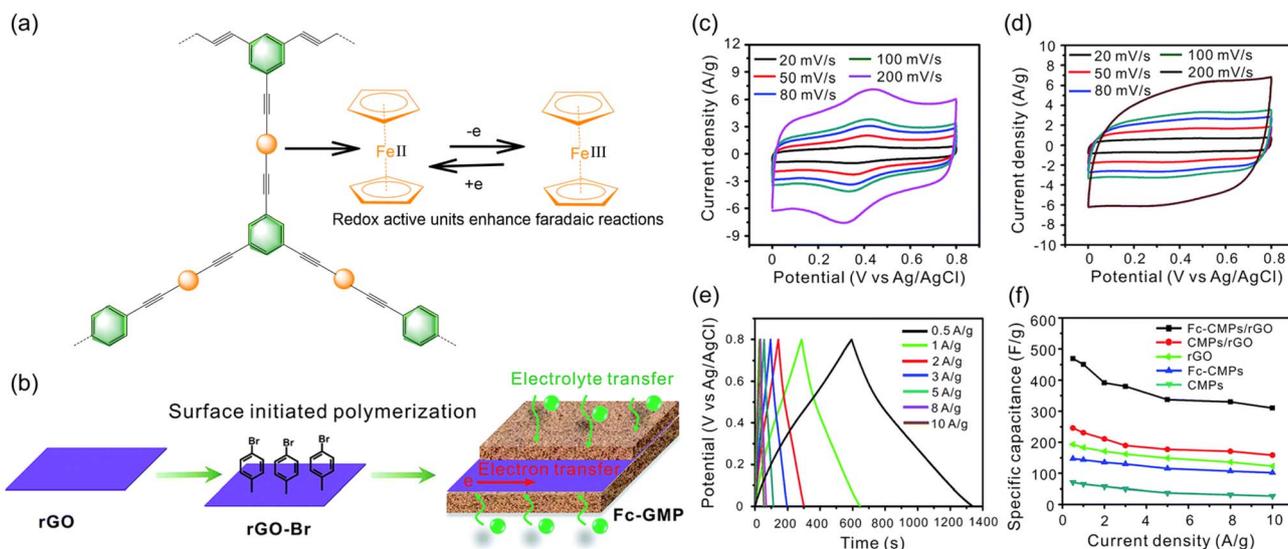


Fig. 18 (a) Chemical structure of redox-active CMPs featuring ferrocene units, along with the corresponding redox reactions of the ferrocene moieties, (b) visual representation of the preparation of Fc-CMPs/rGO hybrids *via* surface-initiated polymerization, (c and d) CV curves of Fc-CMPs/rGO (c) and CMPs/rGO (d) recorded at various scan rates, (e) GCD curves of Fc-CMPs/rGO measured at different current densities, and (f) comparative specific capacitance of Fc-CMPs/rGO, CMPs/rGO, pristine rGO, pure Fc-CMP, and pure CMP electrodes.⁸⁹ Adapted with authorization of Royal Society of Chemistry.



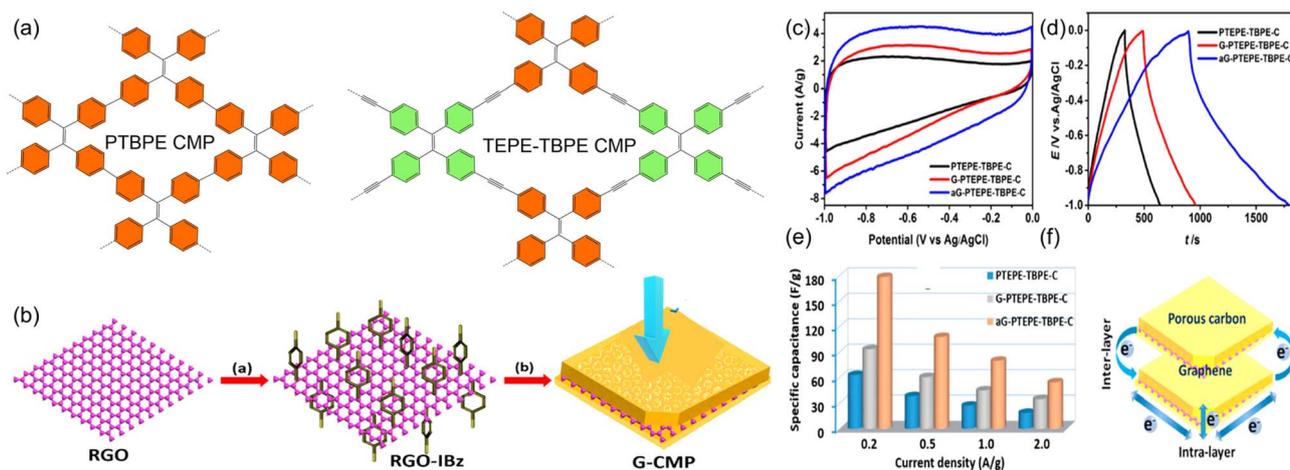


Fig. 19 (a) Chemical structures of PTBPE and PTEPE-TBPE-based CMPs, (b) visual illustration of the design process for graphene-templated CMPs, (c) CV curves of the materials recorded at 100 mV s^{-1} in 6 M KOH aqueous solution, (d) GCD curves measured at a current density of 0.2 A g^{-1} , (e) specific capacitance values at various current densities for supercapacitor devices based on PTEPE-TBPE-C, graphene-templated PTEPE-TBPE-C (G-PTEPE-TBPE-C), and KOH-activated graphene-templated PTEPE-TBPE-C (aG-PTEPE-TBPE-C), (f) schematic representation of the interlayer and intralayer charge transport pathways in the sandwich-like nanosheets of G-PTEPE-TBPE-C during charge-discharge cycles.¹³⁸ Adapted with authorization of American Chemical Society.

conductivity, charge storage, and cycling stability, demonstrating the critical role of SWCNTs in boosting capacitance and energy density for effective supercapacitors.¹¹⁰ The integration of CMPs with multiwalled carbon nanotubes (MWCNTs) *via in situ* polymerization significantly facilitates the capacitance, redox activity, and electrical conductivity of the composite, demonstrating the crucial role of MWCNTs in improving charge transport and energy storage efficiency as shown in Fig. 20. The high surface area ($514 \text{ m}^2 \text{ g}^{-1}$) and interconnected conductive network provided by MWCNTs enhanced rapid ion diffusion as well as electron transfer, resulting in a remarkable capacitance increase of 252% (from 236 F g^{-1} for SACMP to 594 F g^{-1} for MWCNT@SACMP) as well as an energy density of 28.53 Wh kg^{-1} at a power density of 900 W kg^{-1} .¹⁴⁰

5.4 CMP-activated carbon composites

Activated carbon, known for its high surface area and cost-effectiveness, can be combined with CMPs to create composites with balanced performance. The CMPs contribute pseudocapacitance, while activated carbon provides additional double-layer capacitance, resulting in a synergistic enhancement of overall capacitance. Recently, Fe- and N-doped microporous carbon materials were synthesized from Py-BZFC-CMP and Py-PHFC-CMP *via* calcination and KOH activation at $800 \text{ }^\circ\text{C}$. The resulting materials, Py-BZFC-CMP-800 and Py-PHFC-CMP-800, exhibited remarkable properties, including high thermal stability (T_{d10} up to $732 \text{ }^\circ\text{C}$) and substantial char yields (up to $87 \text{ wt}\%$). Moreover, they demonstrated high surface areas up to $376 \text{ m}^2 \text{ g}^{-1}$ and significant pore volumes of $0.36 \text{ cm}^3 \text{ g}^{-1}$, which contribute to enhanced electrochemical performance. Notably, Py-PHFC-CMP-800 achieved an impressive specific capacitance of 324 F g^{-1} at 0.5 A g^{-1} and retained 86% of its capacitance after 5000 charge-discharge cycles at 10 A g^{-1} (Fig. 21). This study highlights how strategic precursor design and activation

processes can transform CMPs into high-performance microporous carbons, where Fe and heteroatom doping significantly enhance their conductivity and charge storage capabilities. The incorporation of activated carbons into CMP-based materials presents a promising avenue for developing efficient energy storage systems with superior electrochemical stability.¹⁴¹ Moreover, Nagai *et al.*¹⁴² demonstrated that activation of COFs, exemplified by nitrogen-rich PI-COF-700, provides a high-surface-area, microporous carbon platform that, when composited with conductive polymers such as PANI and PEDOT/PSS, delivers outstanding capacitance (729.17 F g^{-1}), excellent cycling stability, and superior device performance.

5.5 CMP-metal oxide hybrids

Metal oxides, such as MnO_2 , RuO_2 , and Fe_3O_4 , are renowned for their high theoretical capacitance and redox activity. However, they often suffer from poor cycling stability. Integrating metal oxides with CMPs creates hybrids that combine the advantages of both materials. For CMP- MnO_2 hybrids, MnO_2 is a cost-effective and environmentally friendly pseudocapacitive material.¹⁴³ In CMP- MnO_2 hybrids, the CMPs are expected to provide a conductive and porous framework that supports the MnO_2 nanoparticles, preventing aggregation and enhancing charge transfer. These hybrids may show high specific capacitance as well as excellent cycling retention. In addition, RuO_2 is one of the most promising pseudocapacitive materials due to its high capacitance and conductivity. However, its high cost limits its widespread use. Kumar *et al.*¹⁴⁴ proved that incorporation of Ru/ RuO_2 nanoparticles onto N,S-doped holey graphene significantly enhanced the specific capacitance, demonstrating their effectiveness as active materials for high-performance flexible supercapacitors.

In CMP- RuO_2 hybrids, the CMPs may act as a scaffold that maximizes the utilization of RuO_2 , reducing the amount



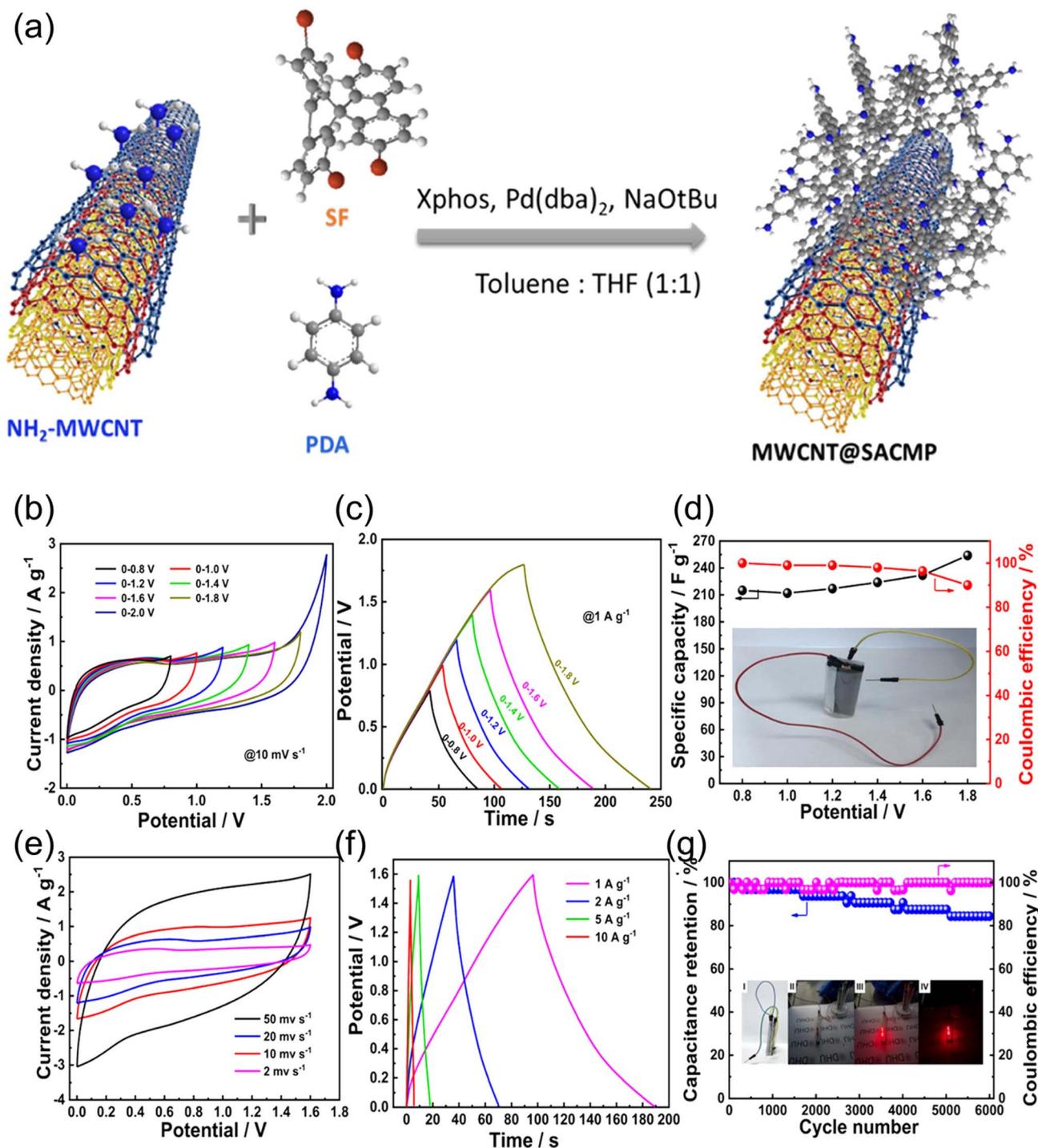


Fig. 20 (a) Schematic illustration of the synthesis of MWCNT@SACMP via the Buchwald–Hartwig (BH) coupling, (b and c) CV and GCD curves of MWCNT@SACMP-3, (d) specific capacitance and coulombic efficiency measured at various operating voltage windows, with the inset displaying a photograph of a single solid-state supercapacitor (SSC) device. (e and f) CV and GCD profiles of MWCNT@SACMP-3 in an operating voltage window of 0–1.6 V, (g) capacitance retention at 2 A g⁻¹ and coulombic efficiency, with insets showing photographs of two SSC devices connected in series: (I) before use, (II) during setup, and (III and IV) after successfully powering LED lights.¹⁴⁰ Adapted with authorization of American Chemical Society.

required while maintaining high performance. These hybrids are especially well-suited for applications requiring high energy density. In addition, Fe₃O₄ offers high redox activity and low

cost.¹⁴⁵ CMP-Fe₃O₄ hybrids leverage the magnetic properties of Fe₃O₄ for easy separation and recycling, while the CMPs provide



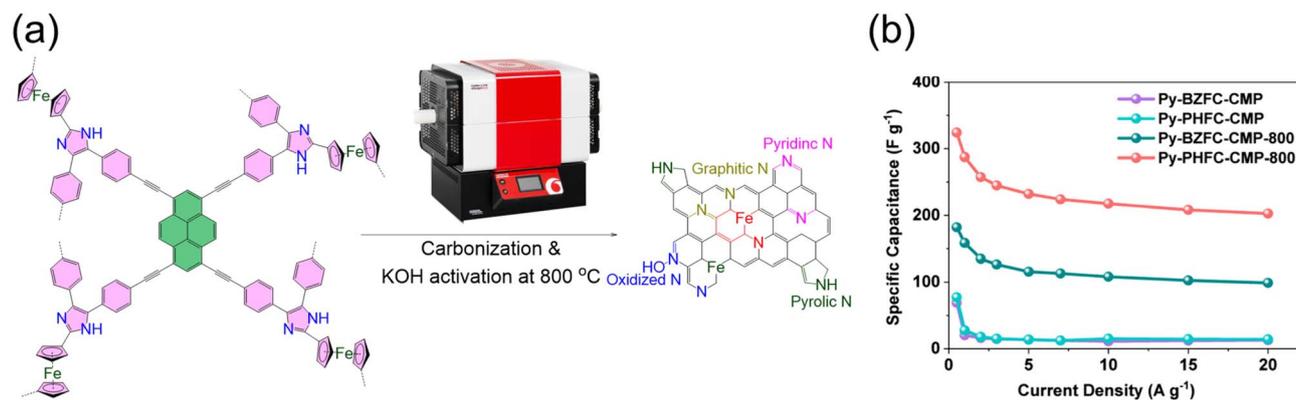


Fig. 21 (a) Schematic illustration of the synthesis of Py-BZFC-CMP-800 from its Py-BZFC-CMP precursor; (b) comparison of specific capacitance values for Py-BZFC-CMP, Py-PHFC-CMP, Py-BZFC-CMP-800, and Py-PHFC-CMP-800.¹⁴¹ Adapted with authorization of American Chemical Society.

structural stability and porosity. These hybrids are promising for environmentally sustainable supercapacitors.

5.6 CMP-conductive polymer blends

Conductive polymers, such as polyaniline (PANI), polypyrrole (PPy), and poly(3,4-ethylenedioxythiophene) (PEDOT), are known for their high conductivity and pseudocapacitive behavior. Blending CMPs with conductive polymers creates composites with enhanced electrochemical performance and processability. PANI is a widely studied conductive polymer due to its high conductivity and ease of synthesis. In CMP-PANI blends, the CMPs provide a rigid and porous framework, while PANI contributes additional pseudocapacitance and conductivity. These blends may exhibit high specific capacitance and excellent rate capability (Fig. 22).¹¹¹ Furthermore, PPy offers high conductivity and environmental stability.¹⁴⁶ CMP-PPy blends combine the microporosity of CMPs with the

redox activity of PPy, resulting in composites with balanced energy and power density. These blends are particularly suitable for flexible and wearable supercapacitors. In addition, PEDOT is known for its high conductivity and stability in oxidized states.¹⁴⁷ CMP-PEDOT blends leverage the synergistic effects of CMPs' porosity and PEDOT's conductivity, rendering them highly suitable for advanced supercapacitor applications.

5.7 CMP-based ternary composites

Ternary composites, which combine CMPs with two other functional materials, offer further performance enhancements by integrating multiple charge storage mechanisms and properties. For example, the composites combine the conductivity of graphene, the pseudocapacitance of metal oxides, and the porosity of CMPs.¹⁴⁸ In addition, a CMP-graphene-MnO₂ ternary composite may show a high specific capacitance, along with excellent cycling retention as well as rate capability.¹⁴⁹

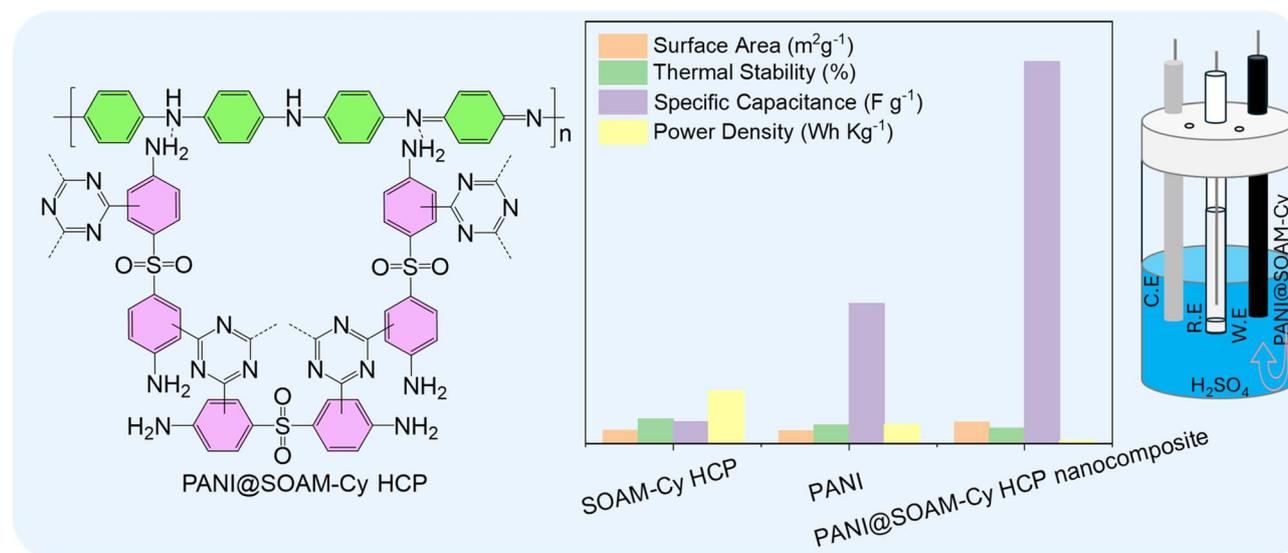


Fig. 22 Molecular structure of the PANI@SOAM-Cy HCP and the influence of polyaniline (PANI) integration on the physical and electrochemical properties of the porous organic polymer (POP).¹¹¹ Adapted with authorization of Elsevier.



Furthermore, the composites integrate the fibrous structure of CNTs and the conductivity offered by conductive polymers, along with the redox functionality of CMPs. They are particularly effective in flexible and high-rate applications.^{20,42,150}

6. Challenges and limitations

While CMPs have shown immense promise as electrode materials for supercapacitors, several challenges and limitations hinder their widespread adoption and practical implementation. These challenges span material synthesis, electrochemical performance, scalability, and device integration. Addressing these issues is critical for advancing CMP-based supercapacitors from laboratory-scale research to real-world applications.

6.1 Material synthesis and design

The synthesis of CMPs often involves multi-step reactions, expensive catalysts (*e.g.*, palladium for Suzuki coupling), and harsh reaction conditions (*e.g.*, high temperatures and inert atmospheres). These factors increase the cost and complexity of production, limiting scalability and commercial viability. In addition, CMPs are inherently porous, and achieving precise control over pore size distribution and hierarchical porosity remains challenging. Irregular pore structures can hinder ion transport and reduce the accessibility of redox-active sites, negatively impacting performance. Furthermore, high porosity is essential for ion adsorption and electrolyte accessibility, but it often comes at the expense of reduced electrical conductivity. Balancing these competing properties is a significant challenge in CMP design.

6.2 Electrochemical performance

Despite their conjugated structures, CMPs generally exhibit lower electrical conductivity compared to traditional carbonous materials (*e.g.*, graphene or activated carbon). This limits their charge transfer kinetics and overall power density. Furthermore, heteroatom doping and functionalization can introduce pseudocapacitance, and the redox activity of CMPs is often insufficient to achieve high energy densities comparable to battery-type materials. Further optimization of redox-active sites is needed. In addition, although CMPs are chemically stable, repeated charge–discharge cycles can lead to structural degradation, particularly in pseudocapacitive CMPs. Ensuring long-term cycling stability is essential for practical applications.

6.3 Scalability and manufacturing

The synthesis of CMPs is typically performed on a small scale in laboratory settings. Scaling up production while maintaining consistent material properties is a significant challenge. In addition, many CMPs are insoluble and infusible, making them difficult to process into thin films or flexible electrodes. Developing processable CMPs or composite systems is crucial for device fabrication. Finally, the high cost of monomers, catalysts, and solvents used in CMP synthesis limits their economic feasibility for large-scale energy storage applications. Developing cost-effective synthetic routes is a priority.

6.4 Device integration and performance

Integrating CMPs into practical supercapacitor devices requires the fabrication of robust and flexible electrodes. However, the brittle nature of many CMPs makes them unsuitable for flexible or wearable applications without additional modifications (*e.g.*, blending with polymers or carbon materials). Also, the performance of CMP-based supercapacitors strongly relies on the choice of electrolyte. Aqueous electrolytes offer high ionic conductivity but limit the operating voltage, while organic and ionic liquid electrolytes offer wider potential windows but may have compatibility issues with CMPs. In addition, CMPs excel in power density due to their rapid charge–discharge capabilities, and their energy density remains lower than that of batteries. Enhancing energy density without compromising power density is a key challenge.

6.5 Characterization and understanding

The complex interplay between EDLC and pseudocapacitance in CMPs is not fully understood. Advanced characterization techniques (*e.g.*, *in situ* spectroscopy and computational modelling) are needed to elucidate these mechanisms deeply. Furthermore, the performance metrics of CMP-based supercapacitors (*e.g.*, capacitance, energy density and cycling stability) are often reported under different testing conditions, making it difficult to compare results across studies. Standardized protocols are essential for meaningful comparisons.

6.6 Environmental and sustainability concerns

Many CMP synthesis routes involve toxic reagents and solvents, raising environmental and safety concerns. Developing greener synthetic methods is critical for sustainable production. Also, the environmental impact of CMP-based supercapacitors at the end of their lifecycle is not well studied. Developing recyclable or biodegradable CMPs is an important consideration for sustainable energy storage.

7. Future directions and opportunities

The field of CMPs for supercapacitor applications is rapidly evolving, with significant potential for breakthroughs in material design, device engineering, and energy storage technologies. While challenges remain, the unique properties of CMPs—such as their tuneable porosity, redox activity, and chemical stability—provide a strong foundation for future advancements.

7.1 Advanced material design and synthesis

Future research should focus on the rational design of CMPs with tailored structures and functionalities. Computational modelling and machine learning can be employed to predict the properties of CMPs and guide the selection of monomers, linkers, and synthetic routes. This approach can accelerate the discovery of high-performance CMPs with optimized porosity, conductivity, and redox activity. Furthermore, developing environmentally friendly and scalable synthesis methods is critical for the commercialization of CMPs. Solvent-free polymerization, mechanochemical synthesis, and the use of inexpensive



catalysts are promising strategies to reduce costs and environmental impact. In addition, designing CMPs with multifunctional properties—such as self-healing, stimuli-responsiveness, or photocatalytic activity—can expand their applications beyond energy storage. For example, self-healing CMPs could enhance the durability of supercapacitors, while photocatalytic CMPs could enable integrated energy conversion and storage systems.

7.2 Enhancing electrochemical performance

Enhancing the electrical conductivity of CMPs is essential for achieving high power density. Strategies such as extended π -conjugation, heteroatom doping, and hybridization with conductive materials should be explored. Moreover, introducing redox-active functional groups or heteroatoms can enhance the pseudocapacitive behaviour of CMPs. Additionally, the integration of CMPs with pseudocapacitive materials (*e.g.*, metal oxides and conductive polymers) can further boost energy density. Furthermore, designing CMPs with hierarchical porosity can optimize ion transport and accessibility, leading to improved rate capability and capacitance. Templating methods and controlled polymerization conditions can be used to achieve this goal.

7.3 Device engineering and integration

Firstly, the advancement of flexible as well as wearable supercapacitors based on CMPs is a promising direction for portable electronics and smart textiles. Fabricating freestanding CMP films, textiles, or composites with high mechanical flexibility and durability is essential for these applications. Secondly, solid-state supercapacitors using gel or solid electrolytes offer improved safety and flexibility compared to liquid electrolytes. Future research should focus on optimizing the compatibility between CMPs and solid electrolytes to enhance performance and stability. Finally, miniaturized supercapacitors based on CMPs are promising for powering microelectronic devices. Modern design techniques, such as inkjet printing, laser scribing, or 3D printing, can be used to create micro-supercapacitors with superior energy and power densities.

7.4. Emerging applications and technologies of CMPs

7.4.1. Integrated energy systems. CMPs can be integrated into hybrid energy systems that combine supercapacitors with batteries, solar cells, or fuel cells. These systems can leverage the high power density of supercapacitors and the efficient energy density of batteries, enabling efficient energy management and storage.

7.4.2. Environmental and sustainable applications. CMPs can be designed for environmentally sustainable applications, such as water purification, CO₂ capture, or energy harvesting.

7.4.3. Smart energy storage. The development of smart supercapacitors with stimuli-responsive properties (*e.g.*, temperature-, pH-, or light-responsive) can enable adaptive energy storage systems. These devices could dynamically adjust their performance based on external conditions, enhancing efficiency and versatility.

8 Conclusions and outlook

CMPs have proven to be a highly promising class of materials for supercapacitor applications, offering an effective integration of large surface area, tunable porosity, redox activity, and chemical stability. Their ability to combine EDLC and pseudocapacitance within a single framework makes them particularly attractive for next-generation energy storage systems. This review has highlighted the structural and chemical features of CMPs, their energy storage mechanisms, recent advances in material design, and the barriers that must be addressed for practical implementation. The most promising strategies identified include: (i) introducing redox-active functional groups and heteroatom doping to enhance charge storage and conductivity; (ii) developing hybrid systems, such as CMP-carbon composites and CMP-metal oxide hybrids, to improve electrochemical performance and durability; (iii) designing hierarchical porosity to facilitate rapid ion transport; and (iv) applying advanced synthetic methods alongside scalable, green approaches to reduce cost and environmental impact. Innovations in device fabrication, including solid-state and micro-supercapacitors, further expand opportunities for CMP integration into portable electronics, smart textiles, and large-scale energy systems. Looking forward, advanced computational tools, machine learning, and standardized testing protocols can accelerate the discovery of high-performance CMPs and promote open collaboration. Moreover, prioritizing recyclable and environmentally friendly CMPs will align their development with global sustainability goals. Importantly, while challenges remain in scalability, conductivity, and processability, the unique properties of CMPs—combined with these promising strategies—offer immense opportunities to advance supercapacitor technology and drive the transition to a fruitful and clean energy future.

Conflicts of interest

There are no conflicts to declare.

Data availability

Data are contained within the article.

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