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Recent Advances in Supercapacitors: Charge-Storage Mechanisms, Electrode Materials, and Future Perspectives

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Abstract. Supercapacitors are valued for rapid power delivery and exceptional cycling endurance, yet increasing the energy stored per unit mass or volume remains difficult. The three representative device configurations in this review are electric double-layer capacitors (EDLCs) based on interfacial ion adsorption, pseudo capacitors that use fast redox or intercalation reactions, and hybrid systems combining both storage modes. Recent progress in the performance of carbon electrodes has focused on creating pores for electrolyte ion access, building continuous transport paths and modifying surface chemistry, rather than increasing the nominal surface area. Redox-active electrodes, such as transition-metal compounds, conjugated polymers, and MXenes and related two-dimensional carbides/nitrides, can store a large amount of charge but often have slow electron transfer kinetics, structural instability and increased interfacial resistance during cycling. A hybrid structure can be used to combine energy storage and power delivery, provided that the two electrodes can operate at the same speed and potential, and the electrolyte is stable in the operating environment. Therefore, future development will move beyond individual material modifications to device-level co-design. The main demands are atomic-level control of the electrode-electrolyte interface, electrolytes with wide electrochemical and thermal operating windows, scalable and sustainable processing, and machine-learning-guided candidate selection. The above optimization is required to develop high-energy-density, long-lasting and manufacturable supercapacitors.

Keywords: Supercapacitors, Electrode materials, Electric double-layer capacitors, Pseudo capacitance, Hybrid supercapacitors

1. Introduction

Pulled by the shift towards renewable power and the escalating energy requirements of terminal users, the advancement of advanced power-storage systems has emerged as a critical necessity. A perfect power-storage framework should concurrently provide superior energy capacity and enhanced power density. Among all types of power-storage devices, supercapacitors have shown good performance in grid frequency regulation, renewable energy integration, regenerative braking systems for high-speed rail, portable electronic devices and electric vehicles due to their high-power density, long cycle life and fast charge/discharge characteristics [1].

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Recently, with the overall goal of improving both energy and power density, many studies have been conducted on nano structural engineering of electrodes, multicomponent assembly, asymmetric or hybrid structures, and flexible and wearable technologies. An excellent example is the modification of the electronic structure of MXenes through a solid-solution method; both the pseudo-capacitance performance has been improved, and at the same time, the development of stretchable fibre-like and flexible supercapacitors has extended their application to wearable electronics and micro-power supplies for the Internet of Things.

Based on the above, this paper will introduce the recent research achievements in the field of supercapacitors. First, this paper will present the general concepts, electrochemical charge-transfer kinetics of electric double-layer capacitance, pseudocapacitance and hybrid energy storage, and explain how all these mechanisms affect the energy- and power-density characteristics of such devices. Next, we will introduce the architectural Design of electrode materials, which includes the optimisation of pore structure and surface functionalisation for porous carbon, nanoarchitecture and synergistic integration of transition-metal oxides with conducting polymers, interfacial and interlayer engineering of advanced materials such as MXenes, etc., all of which can improve the specific capacitance, rate capability and cycling stability of these materials in many ways. Finally, briefly list the problems in the optimisation of energy density, mechanistic understanding, cost control and commercialisation, such as the interfacial compatibility of hybrid structures, scalable production, performance loss at cryogenic temperatures, and the high cost of precious-metal-based electrode materials, and mention some directions for future research as well.

2. Fundamentals and classification

2.1. Fundamentals

A supercapacitor has a positive electrode, a negative electrode, an electrolyte, a separator and current collectors. Electrodes are electrically conductive and have a large surface area for charge accumulation or reaction; the separator is not in direct contact with the two electrodes, and ions in the electrolyte can pass through it; current collectors carry electrons from the electrodes to the outside circuit. Most charge storage happens at the nano-scale electrode/electrolyte interface.

2.2. Classification

The three types of electrochemical capacitors mentioned above have different basic charge-storage mechanisms and are generally classified as electric double-layer capacitors (EDLCs) or hybrid supercapacitors [2]. EDLCs store energy via electrostatic attraction at the junction between the liquid electrolyte and the electrically conductive anode. Cations migrate toward the cathode while anions shift toward the anode under the influence of the applied voltage during the charging process, creating inverse-polarized electric double layers at both electrode-electrolyte boundaries [3]. Since no phase transition or Faradaic redox occurs; it possesses an extremely rapid response time and superior cycling durability; consequently, ideal EDLCs are anticipated to exhibit nearly rectangular CV scans and symmetric triangular GCD waveforms [4].

Energy storage in pseudo capacitors involves fast, reversible Faradaic reactions that occur at or near the electrode interface, such as surface redox reactions, underpotential deposition and intercalation pseudo capacitance. Surface redox and underpotential deposition are still mainly confined to the surface, and intercalation pseudo capacitance involves ion entry into the crystal lattice; although some solid-state diffusion may occur, it is kinetically superior to diffusion-limited

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battery reactions. Therefore, CV plots generally have wide redox regions, and GCD traces may not be in an ideal triangular shape and show potential-dependent gradients [5]. Although these materials have a relatively high specific capacitance compared with carbon-based EDLC electrodes, they generally have problems of conductivity, rate performance and cycling durability.

The majority of pseudocapacitive electrode substances consist of transition-metal oxides, conductive polymers plus novel two-dimensional transition-metal carbides and nitrides. Ruthenium oxide was formerly regarded as a superior candidate owing to its extremely high theoretical specific capacitance and excellent reversibility, yet it remains prohibitively costly for broad application. Manganese dioxide possesses a substantial theoretical specific capacitance, is plentiful and eco-friendly, however, it exhibits poor intrinsic electrical conductivity and requires enhancement via nano structuring plus blending with a conductive agent. Polymers like polyaniline, polypyrrole and poly(3,4-ethylenedioxythiophene) (PEDOT) demonstrate pseudo capacitance through redox doping-dedoping; they are comparatively simple to synthesize and possess good mechanical flexibility, but repeated volume fluctuations over a long duration cause a decline in capacity. MXenes featuring $Ti_3C_2T_x$ have been introduced in recent times and have slowly gained interest because of their integrated attributes of metals plus intercalation pseudo capacitors [6].

The dual electrodes of a hybrid supercapacitor possess distinct energy-retaining processes, and typically comprise a battery-like Faradaic component alongside a capacitive (double-layer or pseudocapacitive) part. A Faradaic electrode retains charge via redox transformations or ion insertion/ deinsertion; it exhibits a specific voltage plateau and a redox peak; a capacitive electrode accumulates charge by means of ion adsorption/desorption plus surface capacitance. The total voltage span, operational voltage and power attributes of the device are defined by the integration of these two electrodes, and a significant boost in energy-storage capacity is attained at high power densities. Since these two mechanisms are inherently dissimilar, the electrode materials for hybrid supercapacitors must differ from those utilized in the aforementioned two types of devices.

3. Electrode materials and research progress

3.1. Electric double-layer capacitors

Electrode substances for EDLCs are primarily carbon-based, with activated carbon serving as the most typical example, possessing specific surface areas surpassing 2000 m^2 g^{-1} and specific capacitances usually spanning 100 to 200 F g^{-1}. Nevertheless, a substantial BET surface area does not ensure superior capacitance: solely ion-accessible surface area provides meaningful contribution, and efficiency further relies on the alignment between pore dimensions and solvated or desolvated ion sizes, pore networking, surface hydrophilicity, and electronic conductivity [7, 8]. Due to their superior electrical conductivity and unique pore architectures, one-dimensional carbon nanotubes plus two-dimensional graphene have emerged as iconic next-generation high-performance carbon materials [9]. Carbon aerogels, ordered mesoporous carbons, and porous carbons synthesized from metal–organic frameworks have likewise garnered significant interest owing to their adjustable pore scales and brief ion-migration routes.

Recent EDLC research has shifted from simply maximizing BET surface area toward coordinating pore size, ion desolvation, and transport pathways. Experiments on carbide-derived carbons showed that subnanometer pores can enhance capacitance when their dimensions approach those of desolvated ions, and systematic ion-size/pore-size comparisons further demonstrated that the optimal pore size depends on the electrolyte [8, 9]. Building on this principle, poly(ionic liquid)-derived carbons engineered for high-voltage electrolytes improved pore–ion compatibility and the

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energy–power trade-off [10], while heteroatom-doped hierarchical porous carbon electrodes combined accessible micro- and mesopores with rapid electron/ion transport at practical mass loadings [11]. Together, these studies indicate that high-performance EDLC electrodes require a balance among ion-accessible micropores, mesoporous transport channels, electrical conductivity, surface chemistry, and practical mass loading, rather than a high nominal surface area alone.

3.2. Pseudo capacitors

Pseudocapacitive devices accumulate charge through rapid and reversible Faradaic redox processes taking place at the interface or near the surface of active phases, involving mechanisms such as surface redox, underpotential deposition, and intercalation pseudo capacitance. Transition-metal oxides were the first pseudocapacitive materials studied to a large extent, and amorphous hydrous ruthenium oxide has achieved a specific capacitance of 720 F g^{-1}, but its rarity and high cost of noble metals have limited widespread application [12]. Manganese dioxide (MnO_2) has attracted much attention because it has a relatively high theoretical specific capacitance of 1370 $F \cdot g^{-1}$ and is abundant and environmentally friendly. MnO_x in water generally has only surface Faradaic reactions, and because it has a very small intrinsic electrical conductivity (10^{-5}-10^{-6} S m^{-1}), the actual specific capacitance is much lower than the theoretical value. Defect engineering, ion pre-intercalation and the construction of low-dimensional nanostructures have all been used to address the problem of conductivity bottleneck [13].

Zhao et al. [14] introduced oxygen defects in MnO_2 nanowires by hydrogen-plasma treatment to increase the carrier concentration by about two orders of magnitude and improve the specific capacitance to 386 F g^{-1} at a current density of 1 A g^{-1}. At the same time, recently, MXenes have been applied to the problem of pseudo-capacitance, and a high-volumetric-capacitance material has been developed via cation intercalation in $Ti_3C_2T_x$. MXenes are easily oxidized in the presence of oxygen in water and therefore have poor cycling stability. Tetra-butylammonium cation pre-intercalation and fluorinated surface termination were used to form a hydrophobic protective layer, and after 15,000 cycles, the capacity retention reached 92%; this is much higher than the 65% retention of untreated MXene, and a new design route for next-generation energy-storage systems has been proposed. Polyaniline, polypyrrole and PEDOT are all conductive polymers that have pseudo-capacitance due to redox reactions in doping-dedoping cycles and possess the advantages of mechanical flexibility and room-temperature processability.

Yao et al. [15] determined that polyaniline has a specific capacitance of 400-600 Fg^{-1}, but after many cycles of swelling and shrinking during galvanostatic cycling, it decomposes and rapidly loses capacity; often, it drops to less than 70% after 1000 cycles. Recently, a large number of problems in polymer/carbon nanocomposite gels have been addressed. In situ synthesis of polyaniline on 3D porous hydrogel electrodes with reduced graphene oxide is carried out to reduce the volume change of the graphene structure and form a stable conductive network; at 10 A g^{-1}, a specific capacitance of 472 F g^{-1} was obtained, and after 2500 cycles, the capacity was still about 91 per cent of the original value, which is much higher than that of traditional pristine polyaniline. Although it has been shown that pseudo-capacitors have a much higher specific capacitance than carbon-based materials and should theoretically have good charge-discharge performance, high capacity and long cycle life remain a problem in practice, and no mature technical path has been found to date.

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3.3. Hybrid supercapacitors

Hybrid supercapacitors (HSCs) combine the high-power density and long cycle life of capacitive electrodes with the high energy density of battery-type electrodes. Battery-type electrodes typically employ transition-metal oxides/hydroxides, sulfides, or insertion-type compounds with high specific capacity—such as $NiCo_2O_4$, $Ni(OH)_2$, Co_3O_4, MoS_2, and $Li_4Ti_5O_{12}$—which provide high energy density, whereas capacitive electrodes generally consist of porous activated carbon, graphene, or other carbon materials, together with small amounts of stable pseudocapacitive materials, ensuring rapid response and high power.

For carbon-based/metal oxide–hydroxide composite electrodes, Ray et al. [16] grew NiCo-layered double hydroxide (LDH) in situ on graphene sheets via a hydrothermal method, yielding a composite electrode with a specific capacitance as high as 1870 F g^{-1} at 1 A g^{-1}, although its rate capability decayed by approximately 35% at 10 A g^{-1}. In recent years, such materials have advanced markedly through interfacial regulation and nanostructure engineering. For instance, a composite of ZIF-67-derived nitrogen-doped porous carbon and Co_9S_8 nanoparticles provides bicontinuous ion/electron channels through an interconnected carbon network with micro- and mesopores; a hybrid capacitor assembled with this material as the battery-type positive electrode and activated carbon as the negative electrode operates at 1.6 V in 1 mol L-1 KOH electrolyte, delivering an energy density of up to 52 Wh kg^{-1} with 82% capacity retention after 10000 cycles.

Advancements have likewise occurred within carbon-based/conducting-polymer hybrid frameworks. Zhu et al. [17] electrodeposited polypyrrole onto ordered carbon-nanotube arrays, utilizing swift electron diffusion via the aligned nanotubes and porous transport pathways; the CV plots maintained a quasi-rectangular profile even as the scan rate was elevated to 50 mV s^{-1}. The integrated all-solid-state hybrid gadget provided an energy density of 25 Wh kg^{-1} at a power density of 850 W kg^{-1}, showing no notable performance decline following 1000 bending iterations. Concerning the pursuit of elevated-voltage aqueous electrolytes, the "water-in-salt" ultra-concentrated electrolyte technique has expanded the aqueous electrochemical window past 3.0 V. Inspired by this method, a hybrid capacitor constructed with Nb_2O_5 nanosheets as the anode, activated carbon as the cathode, and an aqueous LiTFSI electrolyte attained a voltage of 2.8 V and an energy density of 68 Wh kg^{-1}, preserving 65% of its ambient-temperature performance at −30°C and thus illustrating the potential of high-voltage aqueous hybrid systems for broad-temperature functioning [18]. Furthermore, MXene-based hybrid capacitors have garnered increasing focus.

In the course of composite modification, atomic-level interfacial coupling has gained increasing attention. As highlighted by Gowtham et al. [19], an ultrathin (~2 nm) MnO_2 film was conformally coated onto carbon-nanotube surfaces by atomic layer deposition, forming a continuous heterostructure that reduced the interfacial resistance by approximately 60%, with almost no capacity loss after 5000 bending cycles and 10000 charge–discharge cycles. In terms of microstructural control, three-dimensionally ordered hierarchical-pore architectures accelerate ion diffusion at the material surface and interior while mitigating concentration-polarization effects.

Kang et al. [20] increased the mass loading of 3D-printed graphene/iron-oxide composite lattice electrodes to 30 mg cm^{-2}—approaching commercial levels—with a volumetric energy density reaching 12 mWh cm^{-3}. Regarding interfacial engineering, introducing intermediate buffer layers such as graphene oxide or polydopamine between the carbon matrix and metal compounds can alleviate lattice mismatch and enhance mechanical strength, and multiple studies have demonstrated that this strategy can improve the cycle life by more than 50%.

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To overcome poor low-temperature performance, Ma et al. [21] employed an acetamide–water deep-eutectic-solvent electrolyte, which lowers the freezing point while maintaining an ionic conductivity above 0.8 mS cm^{-1} even as the temperature drops to −40 °C. A carbon//Zn hybrid capacitor assembled with this electrolyte retained 82% of its room-temperature energy density and showed no decay after 10000 cycles.

In terms of process evolution, the biomass-carbon route costs less than the carbon-nanotube route, while the introduction of continuous electrospinning and roll-to-roll printing technologies offers new avenues for the large-scale manufacturing of hybrid capacitors. Moreover, machine-learning-assisted materials screening and formulation optimization have emerged as a new research hotspot since 2023 and are expected to substantially shorten development cycles and reduce trial-and-error costs.

4. Current challenges and future perspectives

Although the performance has improved significantly, many problems have not been solved in the process of industrialization. The first is the interfacial compatibility of composite materials: contact between carbon-based materials and metal oxides/hydroxides generally has a high interfacial resistance, and lattice strain and volume expansion during repeated charge-discharge cycles cause the detachment of active materials, leading to a gradual decline in capacity and rate performance. Second, scalable fabrication processes are still immature: most high-performance composite electrodes require complex hydrothermal/solvothermal reactions or chemical vapour deposition, and high-throughput, low-cost, and controllable production have not been realized. In addition, the devices have poor low-temperature performance: aqueous electrolytes readily freeze below 0 °C, and the viscosity of organic electrolytes increases sharply and their ionic conductivity decreases, so the retention of low-temperature capacity is generally less than 50%. Some high-performance battery materials are also based on cobalt and nickel, which are costly and have supply chain risks.

The following are the directions for future study. First, the design and application of two-dimensional materials, such as MBENES, layered double hydroxides and transition-metal chalcogenides, as high-capacity electrodes, along with atomistic-level understanding of interfacial ion transport and structural evolution. Second, screening of electrode/electrolyte combinations using machine learning and high-throughput computation to provide a basis for multi-objective performance trade-offs. Third, the development of all-solid-state and quasi-solid-state hybrid supercapacitors to address the safety and temperature-range limitations of liquid electrolytes, and to enable the large-scale application of flexible and wearable devices. Fourth, green and low-cost synthesis routes based on biomass or recycled feedstocks that meet the requirements of sustainable development and the low-carbon economy. Interfacial science, advanced fabrication technology and artificial intelligence are now being combined to expand the applications of supercapacitors in electric vehicles, intelligent grids, smart devices and other specialized equipment.

5. Conclusions

Due to their inherent benefits of superior power density and extended cycle life, supercapacitors have emerged as a vital element within electrochemical energy-storage architectures. This article has comprehensively examined the operational principles, material frameworks, and electrochemical performance attributes of three device types—EDLCs, pseudo capacitors, and hybrid supercapacitors. EDLCs stand out for their high-power delivery and superior cycling longevity and maintain technological primacy in high-rate charge–discharge environments; pseudo capacitors

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significantly boost specific capacitance via rapid and reversible Faradaic redox processes, though their rate performance and long-term cycling robustness require additional refinement; and hybrid supercapacitors, by merging the kinetic features of non-Faradaic and Faradaic electrodes, represent the primary technical trajectory toward balancing high energy density with high power density. In recent times, thanks to heteroatom-doped carbon materials, defect-modified transition-metal oxides, in situ composite heterostructures, and highly concentrated aqueous and organic electrolytes, hybrid devices have reduced the disparity under laboratory evaluation criteria in material synthesis and interface engineering, with energy densities reaching those of lithium-ion batteries while preserving outstanding power traits and enduring cycling stability.

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