REVIEW ARTICLE

SPINEL-BASED MATERIALS FOR HIGH-PERFORMANCE SUPERCAPACITORS - A REVIEW ARTICLE

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Abstract. *Spinel metal oxides (general formula* AB_2O_4*) have received strong research attention as electrode materials for supercapacitors because of their rich redox activity, stable structure, and low cost [1]. Their unique crystal arrangement, where metal ions sit in tetrahedral and octahedral sites, allows easy tuning of electrical, magnetic, and electrochemical behavior. In recent years, many spinels such as* $NiCo_2O_4$, $MnCo_2O_4$, $ZnFe_2O_4$, Co_3O_4, $FeCo_2O_4$, *and high-entropy spinels have been studied for fast charge storage. Researchers have shown that controlling the synthesis method, morphology, porosity, and particle size can significantly improve their capacitance, cycling stability, and rate performance. Spinel-carbon composites, conducting polymer hybrids, and asymmetric devices further enhance practical device performance. However, challenges remain, including moderate conductivity, structural degradation during cycling, and difficulties in achieving high mass loading for real devices [2]. This review summarizes the fundamentals of spinel structure, recent progress in synthesis and nanostructure design, electrochemical mechanisms, device-level performance, challenges, and future directions. The goal is to provide a clear and practical understanding of how spinels can contribute to next-generation, high-efficiency.*

Keywords: spinel oxides; supercapacitors; pseudocapacitive materials; nanostructured electrodes; energy storage review.

1. Introduction

The global demand for efficient energy storage technologies is rapidly increasing as renewable energy deployment, electric transportation, and portable electronics continue to expand worldwide [3]. Renewable sources such as solar and wind are clean but intermittent, meaning that harvested energy must be stored and supplied on demand to maintain grid stability and efficiency. Likewise, electric vehicles require energy-storage devices that can rapidly charge and release energy, delivering high power during acceleration and regenerative braking. In this context, supercapacitors, also called electrochemical capacitors, have emerged as highly promising devices because they store and release charge through fast surface redox reactions and rapid ion transport rather than slow bulk chemical processes typical of batteries. As a result, supercapacitors offer exceptional features including extremely high power density, fast charge-discharge capability, outstanding cycling stability, long operational life, and inherent safety, making them suitable for applications such as hybrid vehicles, emergency power backup, industrial power smoothing, and wearable electronics [4-6]. However, the major limitation that still restricts large-scale adoption is their comparatively low energy density. While supercapacitors can release energy very rapidly, their total stored energy remains lower than that of lithium-ion batteries. Therefore, improving the energy density of supercapacitors has become one of the most active directions in modern energy-storage research [7], and progress strongly depends on the development of

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advanced electrode materials capable of combining high capacitance, fast electrochemical kinetics, and long-term stability [8].

Among the many electrode materials investigated, spinel metal oxides with the general formula AB_2O_4 have received significant attention in recent years [9]. Numerous studies have highlighted the outstanding electrochemical behavior of spinel oxides such as $NiCo_2O_4$, Co_3O_4, $MnCo_2O_4$, $ZnMn_2O_4$, and Fe_3O_4 for high-performance supercapacitors [4,10-13]. These materials possess a three-dimensional face-centered cubic oxygen framework, where metal cations occupy both tetrahedral (A-site) and octahedral (B-site) crystal positions. This arrangement creates a robust and stable lattice capable of tolerating repeated ion insertion and extraction during cycling without structural collapse. Another major advantage of spinel oxides is the presence of multivalent metal ions, such as Ni^{2+}/Ni^{3+}, Co^{2+}/Co^{3+}, $Mn^{2+}/Mn^{3+}/Mn^{4+}$, and Fe^{2+}/Fe^{3+}, which provide multiple reversible redox pathways that contribute to strong pseudocapacitance and higher charge-storage capability than purely double-layer carbons. Recent works between 2024 and 2025 have demonstrated that nanostructured spinel electrodes can achieve very high capacitance values-often exceeding 1500 $F.g^{-1}$-when engineered into nanosheets, hollow spheres, and hierarchical porous structures, owing to improved electronic conductivity and accessible redox sites [11-14]. Spinel ferrites, including $NiFe_2O_4$ and $CoFe_2O_4$, have also shown excellent cycling stability, particularly when combined with conductive matrices such as graphene, carbon nanotubes, MXenes, and conductive polymers, which enhance electron transport and mitigate mechanical degradation during long-term cycling [10-12].

The goal of this review is to provide a clear, accessible, and well-organized discussion of recent progress in spinel-based materials for supercapacitors, written in a simplified and human-friendly style without losing scientific depth. The review first explains the structural characteristics and chemical diversity of spinel oxides that make them suitable for pseudocapacitive charge storage, followed by a detailed overview of the major spinel families, including cobalt-based, nickel-based, manganese-based, ferrite, zinc-based, and multicomponent systems, discussing their advantages, limitations, and performance trends. Various synthesis methods-such as hydrothermal synthesis, co-precipitation, solvothermal methods, electrospinning, microwave processing, spray pyrolysis, and template-assisted routes-are outlined, since they strongly influence the morphology, conductivity, and surface area of the resulting electrode materials [1]. The review also summarizes recent electrochemical performance achievements, analyzing key parameters such as specific capacitance, rate capability, ion diffusion, charge-transfer resistance, coulombic efficiency, and long-term durability. Mechanistic aspects, including redox processes, ion-transport pathways, electron conduction, and structural stability, are also discussed to provide a deep understanding of how spinel oxides function during repeated charge-discharge cycles. Finally, key challenges facing spinel-based materials-such as limited intrinsic conductivity, structural degradation at high current density, and difficulties in large-scale production-are addressed, and future research directions are proposed, including doping strategies, defect engineering, hybrid composites, multimetal substitution, and the development of flexible and solid-state supercapacitor devices. Through this comprehensive overview, the review aims to support both new researchers entering the field and experienced scientists working toward the rational design of next-generation spinel-based electrodes for high-performance energy-storage systems.

2. Background on Spinel Structure

Spinel is a versatile class of crystalline materials with the general formula AB_2O_4 (Fig.1), characterized by a well-defined three-dimensional cubic close-packed oxygen lattice that provides a stable and flexible framework for incorporating a wide range of metal cations, making it an important structural platform for electrochemical applications such as supercapacitors. In the typical spinel configuration, A-site cations occupy one-eighth of the available tetrahedral interstices, while B-site cations fill half of the octahedral interstices, though the exact cation distribution can vary depending on the electronic structure, ionic radii, electronegativity, and formation environment of the constituent metals [15]. Spinels are traditionally classified into three main types-normal spinels, inverse spinels, and mixed (partially inverse) spinels-according to the cation arrangement across tetrahedral and octahedral lattice sites; for example, in normal spinels such as $ZnAl_2O_4$, divalent A-site cations reside in tetrahedral positions and trivalent B-site cations in octahedral ones, whereas in inverse spinels like Fe_3O_4, half of the trivalent cations move to tetrahedral sites while the divalent and remaining trivalent ions share the octahedral sites [16]. This unique distribution results in rich defect chemistry and high variability in electronic properties, allowing spinels to exhibit metallic, semiconducting, or insulating behavior depending on the degree of cation ordering, electron hopping pathways, and charge compensation mechanisms. The presence of multivalent transition metals such as Mn, Co, Ni, and Fe further enhances redox activity, creating abundant energy storage sites that make spinel-based oxides particularly attractive for faradaic charge storage in supercapacitor electrodes, where reversible ion-electron transfer occurs during cycling [17]. Additionally, the robust cubic structure imparts high mechanical stability, thermal resistance, and tolerance to volume changes, allowing the materials to withstand repetitive redox cycling without structural degradation. Spinels also offer a large surface area when engineered into nanoscale morphologies, providing short ion diffusion distances and optimized electrolyte contact. Therefore, the spinel structure effectively combines structural integrity, chemical versatility, electronic tunability, and electrochemical responsiveness, explaining why AB_2O_4 oxides such as $MnCo_2O_4$, $NiCo_2O_4$, $CuCo_2O_4$, and $MnFe_2O_4$ have emerged as leading electrode materials in next-generation high-performance pseudocapacitors [18,19].

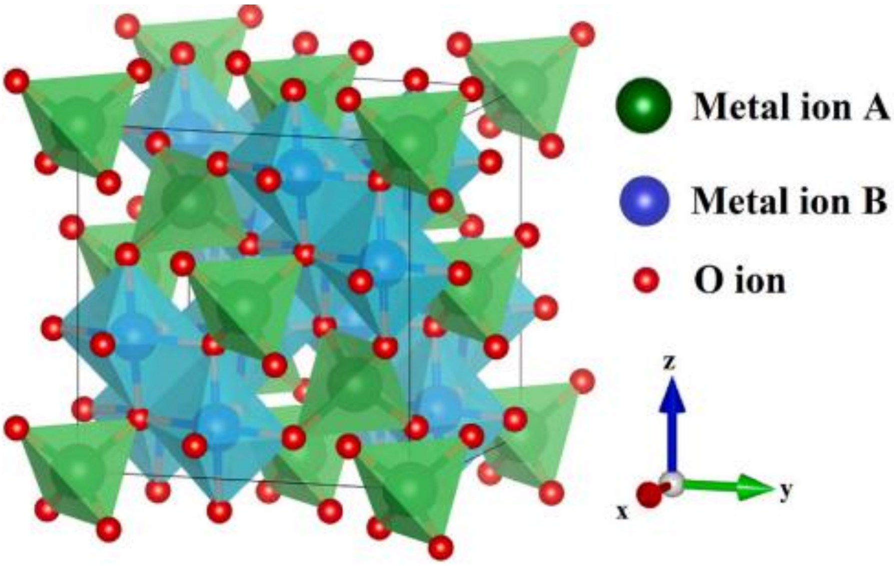

Fig. 1. AB_2O_4 spinel oxide structure [20].

3. Why Spinels Are Attractive for Supercapacitors

Spinel metal oxides (AB_2O_4) have become some of the most promising and widely studied electrode materials for supercapacitors because they combine several key characteristics that directly enhance charge storage performance and long-term stability [7]. One of the most important reasons for their strong performance is their rich redox activity. Unlike pure carbon materials, which store energy mainly through electrostatic ion adsorption, spinels contain transition metals such as Ni, Co, Mn, and Fe that can exist in multiple oxidation states [21], allowing them to participate in fast and reversible faradaic reactions. These redox processes significantly increase the amount of charge stored per unit mass, giving spinel-based electrodes much higher specific capacitance than traditional EDLC systems [22]. For example, materials like $NiCo_2O_4$ can undergo Ni^{2+}/Ni^{3+} and Co^{2+}/Co^{3+} transitions during cycling, which adds multiple electron-transfer pathways to boost pseudocapacitive behavior. Another important advantage is their higher electrical conductivity compared to many simple oxides. Binary oxides like Co_3O_4, NiO, or MnO_2 often suffer from limited conductivity, which restricts electron transport and reduces rate performance. In contrast, bimetallic spinels, particularly $NiCo_2O_4$, are known for their enhanced conductivity due to synergistic interactions between the two metal ions, which create additional charge carriers and facilitate faster electron movement within the lattice. This lower internal resistance helps the electrode deliver high power density and maintain stable operation even at high current loads. In addition to their electrochemical benefits, spinels also demonstrate excellent structural stability, which is essential for devices expected to withstand tens of thousands of charge-discharge cycles. The spinel structure has a robust three-dimensional framework where oxygen anions form a tightly packed cubic lattice, and metal cations occupy tetrahedral (A) and octahedral (B) sites. This well-organized arrangement can accommodate repeated ion insertion and extraction without severe distortion or collapse, making spinel materials much more resistant to mechanical and chemical degradation than many layered or amorphous oxides. Another appealing feature of spinels is their low cost and natural abundance, which is critical for the practical deployment of energy storage technologies. Elements like manganese, iron, cobalt, zinc, and nickel are relatively inexpensive and readily available, allowing the fabrication of low-cost electrode materials compared to noble-metal-based systems. Furthermore, spinels are highly tunable materials, which means their properties can be easily engineered for improved performance. They can be doped with other metal ions to modify electronic structure, substituted at the A or B sites to enhance conductivity or redox activity, and combined with conductive additives such as graphene, reduced graphene oxide, carbon nanotubes, or MXenes to overcome limitations in electron transport. This chemical flexibility allows researchers to design composite or hybrid materials with synergistic effects, resulting in improved mechanical strength, higher surface area, and more accessible active sites. Beyond chemical tuning, spinels can be synthesized into a wide range of nanostructures, including nanosheets, nanowires, nanoneedles, hollow spheres, nanocubes, core–shell structures, and hierarchical porous networks. These engineered shapes not only increase the surface area available for redox reactions but also shorten ion diffusion paths, enhance electrolyte penetration, and improve contact between the active material and the current collector [17]. This combination of intrinsic redox chemistry, good conductivity, structural robustness, low cost, compositional tunability, and versatility in morphology makes spinel oxides exceptionally well-suited for high-performance supercapacitors. As a result, they have become one of the leading classes of pseudocapacitive materials in recent research and continue to attract growing attention for both academic study and real-world energy storage applications.

3.1. Types of Spinels Used in Supercapacitors

Spinel oxides used in supercapacitors cover a wide family of materials (Fig. 2), and each type brings unique electrochemical advantages because of its cation configuration, redox activity, and conductivity. Among them, cobalt-based spinels [23] such as Co_3O_4 and $NiCo_2O_4$ are the most widely studied due to their high electrical conductivity and rich redox chemistry [24]. Co_3O_4 itself has good pseudocapacitance but suffers from moderate conductivity; however, when cobalt is combined with nickel to form $NiCo_2O_4$, the resulting bimetallic spinel exhibits superior performance because Ni^{2+}/Ni^{3+} and Co^{2+}/Co^{3+} pairs offer multiple electron-transfer pathways. This synergy also improves conductivity significantly, making $NiCo_2O_4$ one of the highest-performing spinel materials for supercapacitor electrodes. Another important family is manganese-based spinels [25], including $MnCo_2O_4$, $ZnMn_2O_4$, $NiMn_2O_4$, and $MnFe_2O_4$. Manganese has multiple oxidation states ($Mn^{2+}/Mn^{3+}/Mn^{4+}$), which provide abundant active sites for faradaic reactions. Manganese spinels are also attractive because manganese is low-cost, environmentally friendly, and available in large quantities. For example, $MnCo_2O_4$ often displays excellent cycling life due to manganese's structural stability, and $ZnMn_2O_4$ can offer high specific capacitance thanks to its mixed cation distribution, which improves electron hopping and ion diffusion. Nickel-based spinels [26], such as $NiFe_2O_4$ and $NiMn_2O_4$, are also promising. Nickel ferrite ($NiFe_2O_4$), for instance, has shown strong pseudocapacitive behavior due to the Fe^{2+}/Fe^{3+} redox pair and the Ni^{2+}/Ni^{3+} couple. Although ferrites generally have lower conductivity than cobaltites, this limitation can be overcome through compositing with conductive materials such as graphene or carbon nanotubes, leading to impressive energy density in hybrid supercapacitors [27]. Iron-based spinels [28], especially Fe_3O_4 and $CoFe_2O_4$, offer good electrochemical activity combined with very low cost and environmental safety. Fe_3O_4 is magnetic and provides a high theoretical capacitance, but it can suffer from cycling instability unless combined with carbon or engineered into stable nanostructures. $CoFe_2O_4$ has better conductivity and mechanical strength, making it more suitable for long-term cycling applications. Another useful group includes zinc-based spinels [29] like $ZnCo_2O_4$ and $ZnMn_2O_4$, which are known for their high porosity, abundant oxygen vacancies, and relatively low cost. Zinc also helps stabilize the spinel framework, reducing degradation during repeated charge–discharge cycles. $ZnCo_2O_4$ in particular has shown excellent rate performance in many studies because cobalt enhances the overall conductivity while zinc improves structural stability. Beyond these traditional systems, multicomponent spinels [31] containing three or more metal ions-such as $NiCuCo_2O_4$, $MnCoFe_2O_4$, or $ZnNiCo_2O_4$-are attracting growing attention. These materials combine the advantages of multiple metal ions, creating richer redox chemistry and improved conductivity through synergistic effects. For example, Ni-Cu-Co spinels often show better electron transport and superior capacitance compared to their binary counterparts. Researchers are also developing doped spinels [30] where small amounts of metal ions like Al^{3+}, Cr^{3+}, Mg^{2+}, or rare-earth elements are introduced into the lattice to tune conductivity, surface activity, and defect concentration. Doping can increase oxygen vacancies, enhance electron hopping, or strengthen structural stability, leading to improved long-term cycling performance. Overall, the wide variety of spinel oxides-including cobaltites, manganese-based spinels, ferrites, nickel-based spinels, zinc-containing systems, and multimetal/doped spinels-provides a rich library of materials for designing high-performance supercapacitors. Their tunable composition allows researchers to precisely adjust redox activity, conductivity, and morphology, making spinels one of the most flexible and powerful material families in the field of energy storage.

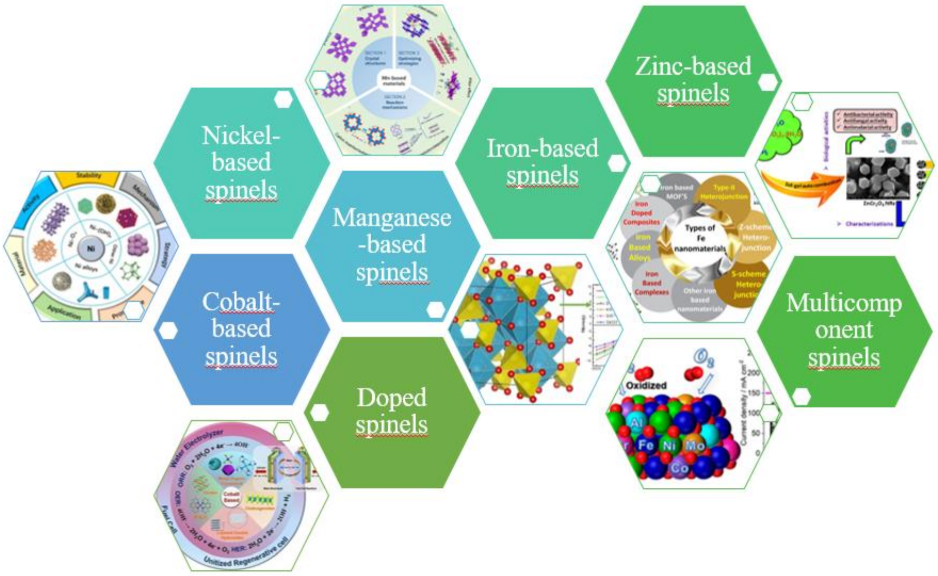

Fig. 2. Overview of different types of spinel materials used in supercapacitors, including cobalt-based, nickel-based, manganese-based, iron-based, zinc-based, doped, and multicomponent spinels, highlighting their structural diversity and potential applications.

3.2. Synthesis Methods

The choice of synthesis method directly governs the morphology, crystallinity, specific surface area, and electrochemical performance of spinel oxides, which is why a wide range of well-established techniques are currently used for (Fig. 3) supercapacitor electrodes: the most popular are hydrothermal and solvothermal routes, which provide precise control over particle size and shape under relatively mild conditions, enabling the fabrication of nanowires, nanosheets, hollow spheres, and hierarchical architectures with high accessible surface area [32,33]; a simple and easily scalable approach remains co-precipitation, in which a mixed hydroxide precursor is precipitated from metal salt solutions and then calcined; excellent compositional homogeneity is achieved via the sol-gel method; one-dimensional fibers are conveniently produced by electrospinning; spherical and hollow particles are obtained through spray pyrolysis; complex porous and core-shell structures are realized using soft or hard template-assisted methods; and when ultra-fast synthesis is required, microwave-assisted routes yield highly crystalline nanoparticles in minutes [34], while combustion synthesis and thermal decomposition can produce fine powders in seconds, albeit with less morphology control [35]. Ultimately, each method offers its own trade-off between structural control, cost, and scalability, and the choice depends on the target shape, conductivity, and electrochemical activity required from the final electrode.

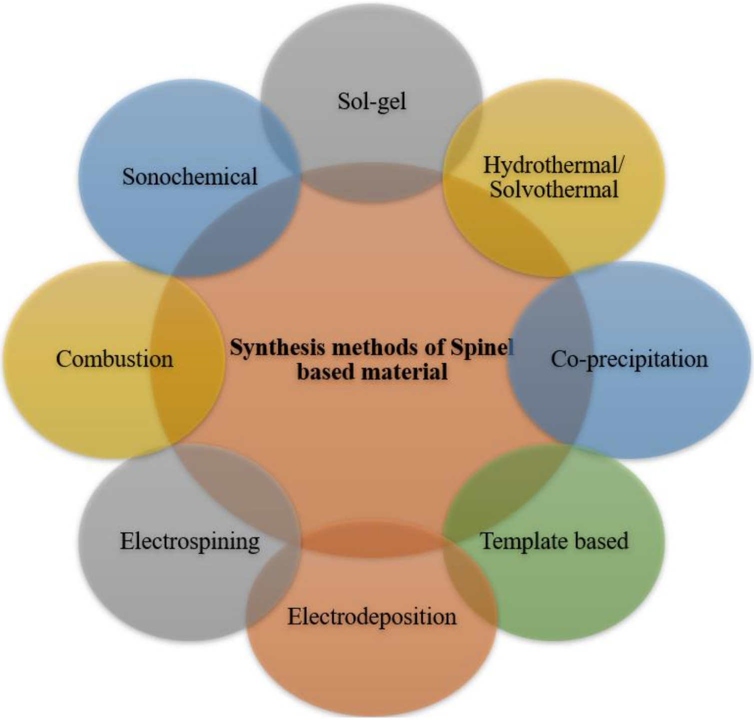

Fig. 3. Techniques involved in the synthesis of spinel-based material.

3.3. Morphologies and nanostructures

The way a spinel material looks at the nanoscale (Fig. 4) (wires, sheets, hollow balls, etc.) basically decides how well it will work in a supercapacitor: the more open the structure, the easier electrolyte to get inside, the shorter the paths for ions, and the faster electrons fly, all while the electrode doesn't fall apart after thousands of cycles. That's why everyone is trying to move away from simple bulky particles and make fancy nano-shapes: 1D nanowires and nanofibers give a highway for electrons straight along their length, 2D nanosheets and flakes expose tons of active surface and cut ion diffusion distance to almost nothing (especially when they're grown directly on nickel foam or carbon cloth so you don't need any binder), and 3D hierarchical stuff (flowers, urchins, nanosheets on wires) combines huge surface area with big open pores so the electrolyte soaks everything instantly, delivering sky-high capacitance even at crazy charging speeds [36,37]. Hollow spheres, core-shell, yolk-shell, and other porous designs take it further: they buffer mechanical stress during cycling and let the electrolyte attack the material from inside as well as outside. Simple nanoparticles and cubes are still around because they pack nicely, but they usually lag behind the more open architectures in rate performance. Bottom line: smart nanostructuring is the fastest and most reliable way to squeeze way more energy, power, and lifetime out of spinel electrodes compared to plain bulk powders.

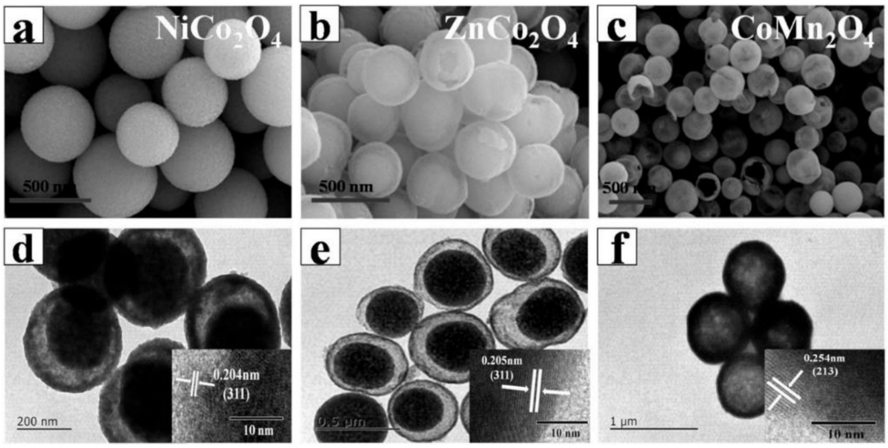

Fig. 4. SEM, TEM, and HRTEM of hollow $NiCo_2O_4$ (a, d), $ZnCo_2O_4$ (b, e), and $CoMn_2O_4$ (c, f) spheres [38,39].

3.4. Electrochemical Performance

Spinel electrodes rock in supercapacitors because they deliver real faradaic punch without sacrificing too much speed or lifetime: you typically see quasi-rectangular CV curves with fat redox peaks, specific capacitance from 800 up to 2000+ F g^{-1} at low currents, and after 5000-10000 cycles they still keep 85-95% of that capacity thanks to their tough 3D lattice that doesn't crumble when the material keeps expanding and shrinking. Charge-discharge curves are nicely sloped (classic pseudocapacitor style), rate capability stays strong even at 20-30 A g^{-1} if the nanostructure is open and conductive (especially with $NiCo_2O_4$ and $MnCo_2O_4$), and EIS shows tiny charge-transfer resistance plus almost vertical lines in the low-frequency zone, meaning ions and electrons fly fast. Put them into real asymmetric or hybrid devices, and you easily hit 40-60 Wh kg^{-1} energy density while still having power like a carbon supercapacitor [40]. Sure, some spinels are still a bit sluggish in pure form and can lose inner surface if the particles are too thick, but that's why everyone now mixes them with graphene, CNTs, or polymers, and the numbers just keep getting better.

3.5. Mechanisms of Charge Storage

The charge storage mechanisms in spinel-based supercapacitors are fundamentally governed by pseudocapacitive Faradaic reactions involving reversible electron transfer between transition-metal ions with multiple oxidation states, allowing significantly higher capacitance than purely electrostatic electric double-layer capacitors (EDLCs). The general spinel structure AB_2O_4 contains A-site cations in tetrahedral positions and B-site cations in octahedral positions, and both can host transition metals such as Ni, Co, Mn, Fe, Cu, and Zn [41], each capable of switching between oxidation states like Ni^{2+}/Ni^{3+}, Co^{2+}/Co^{3+}, Mn^{3+}/Mn^{4+}, or Fe^{2+}/Fe^{3+} during charge–discharge cycling, meaning multiple electrons per formula unit can participate in energy storage. Because of this rich redox chemistry, spinels rely mainly on surface and near-surface pseudocapacitive mechanisms (Fig. 5) where ions from the electrolyte, typically OH^- in alkaline solutions, rapidly adsorb and react at active metal sites according to reactions such as $NiCo_2O_4 + OH^- \rightleftharpoons NiOOH + CoOOH + e^-$, and the reversibility of these reactions ensures high coulombic efficiency and long cycle life. In addition to surface reactions, some spinel materials also support intercalation-based storage, where small ions like H^+, OH^-, or Na^+ insert into the crystal lattice without collapsing the 3D framework. This is possible because the spinel structure is mechanically stable and has spacious ion diffusion channels, allowing ion insertion and extraction with minimal strain, which enables higher energy density resembling battery-type electrodes while retaining fast kinetics suitable for supercapacitors. Electronic conductivity is another factor that enhances charge storage, because the mixed valence states of transition metals in spinels allow electron hopping between neighboring metal ions, making materials such as $NiCo_2O_4$ far more conductive than single-metal oxides like NiO or Co_3O_4. This conductivity ensures faster charge transfer and lower internal resistance, which translates into higher power density. Furthermore, the efficiency of the charge storage mechanism strongly depends on morphology and particle size [40]; nanosheets, nanowires, hollow structures, and porous frameworks greatly increase accessible surface area, reduce diffusion length for ions, expose more active metal sites, and make Faradaic reactions more rapid and complete. Tailoring the composition of spinels also enhances the mechanisms: doping with elements such as Cu, Fe, V, or Cr modifies the electronic structure, increases the number of redox centers, and lowers charge-transfer resistance, while combining spinels with conductive networks like graphene, carbon nanotubes, MXenes, or conductive polymers allows electrons to move quickly throughout the electrode and prevents particle aggregation. These hybrid configurations create multiple interconnected pathways for ion and electron transport, enabling higher specific capacitance, better rate capability, and improved cycling stability [8]. Ultimately, the charge storage behavior of spinel-based electrodes is the result of a synergistic combination of mixed-valence redox activity, fast ion diffusion channels, high electrical conductivity, and structural robustness. And by optimizing crystal chemistry, morphology, and composite design, researchers can tune spinels to balance high energy density, power density, and durability in next-generation supercapacitors.

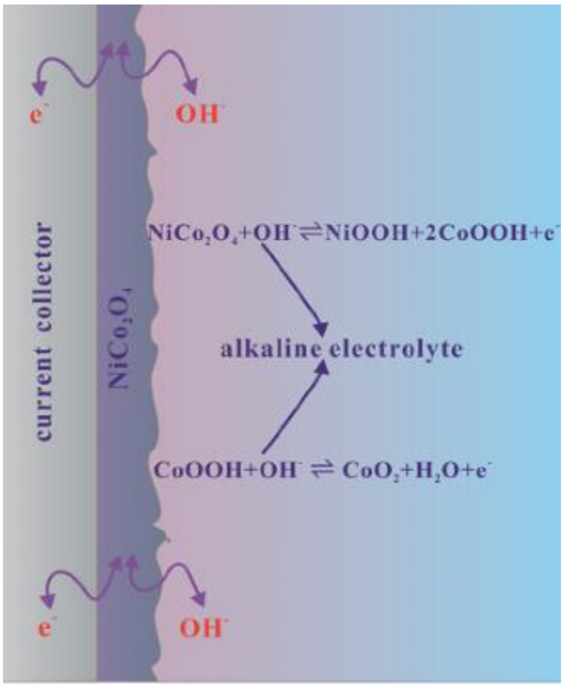

Fig. 5. A schematic of the charge storage mechanism of $NiCo_2O_4$ [40].

3.6. Composite and Hybrid Spinel Materials

Pure spinels are cool on paper with all their redox tricks, but in real life, they often choke on slow conductivity, particles stick together, and the structure slowly crumbles after a few thousand cycles-so the fix everyone uses now is to team them up with something that conducts like crazy and keeps everything in place. Throw graphene, CNTs, MXene, or carbon nanofibers in the mix, and you instantly get a super-highway for electrons, way less resistance, and the spinel nanoparticles stay nicely separated, so every redox site actually works. Add conducting polymers like PANI or PPy, and you get flexible electrodes that bend without breaking, plus extra pseudocapacitance on top. People also love making spinel-on-spinel combos ($NiCo_2O_4$ with $MnCo_2O_4$, $ZnCo_2O_4$ with Fe_3O_4, etc.) because each brings its own strength to the party-one gives huge capacity, the other keeps the juice flowing fast. All these hybrids are built as hollow balls, core shells, cages, or mesoporous jungles, so ions barely have to travel, and mechanical stress gets soaked up instead of cracking the material. The result? Capacitance jumps, rate capability stays solid even at brutal currents, and cycling life easily crosses 10-20k cycles without sweating. Bottom line: pure spinels are yesterday; today's winners are smart composites that take the redox power of spinels and turbo-charge it with conductive scaffolding for real-world, high-energy, high-power, long-life supercaps.

3.7. Device Configurations and Practical Supercapacitors

In practical applications, the performance of spinel-based materials depends not only on their intrinsic electrochemical properties but also on how they are integrated into real device configurations, which include electrode assembly, current collectors, separators, electrolytes, and packaging. Supercapacitors using spinel electrodes are commonly constructed in three main configurations: three-electrode systems, used primarily in laboratory testing to accurately evaluate electrode behavior; two-electrode coin or Swagelok-type cells, which more closely represent real device performance; and full packaged devices such as pouch cells, cylindrical cells, and flexible or wearable architectures designed for consumer electronics and portable energy storage. In laboratory research, the three-electrode setup-consisting of a working electrode coated with the spinel material, a reference electrode such as Ag/AgCl or Hg/HgO, and a counter electrode like platinum wire-allows precise measurement of redox behavior, specific capacitance, and kinetics without interference from other cell components [44]. However, real-world performance is better reflected in two-electrode systems where both positive and negative electrodes operate simultaneously, and losses related to internal resistance, electrode balancing, and charge-transfer efficiency are more accurately captured. For asymmetric supercapacitors, spinels are often employed as positive electrodes due to their pseudocapacitive behavior and high theoretical capacitance, paired with carbon-based negative electrodes providing fast EDLC-type charge storage, resulting in devices that achieve higher operating voltage and improved energy density compared to symmetric configurations. The choice of electrolyte also significantly influences device performance, with aqueous electrolytes (KOH, Na_2SO_4, Li_2SO_4) offering high ionic conductivity and low cost but limited voltage window (typically <1.8 V), while organic or ionic liquid electrolytes allow broader operating voltages of 2.5-3.5 V and greater overall energy density, though at higher cost and increased safety considerations. The interface between the spinel electrode and current collector (often stainless steel, nickel foam, titanium foil, or conductive carbon cloth) must ensure strong adhesion and minimal contact resistance; using 3D conductive scaffolds like nickel foam or carbon cloth provides mechanical support and fast electron transport, improving rate capability. For flexible and wearable energy-storage systems, spinels are increasingly deposited onto polymer substrates, textiles, carbon fibers, or hydrogels to create bendable, stretchable, and lightweight devices suitable for sensors, smart textiles, and portable electronics. Inkjet printing, spray coating, electrophoretic deposition, and roll-to-roll processing are emerging as scalable fabrication methods that support industrial production. Moreover, device-level packaging must ensure stability against electrolyte leakage, corrosion, and mechanical stress, especially for long-term cycling. In performance terms, practical cells using spinel-based electrodes can achieve specific capacitances exceeding 100-200 F g^{-1} in a two-electrode configuration, with areal and volumetric capacitances optimized for space-limited applications. Many systems also demonstrate long life spans, retaining more than 80-95% of initial capacitance after thousands of cycles [1]. As spinel synthesis, electrode design, and device engineering continue to improve, these materials are moving closer to commercial viability, offering high-power, fast-charging, and environmentally friendly energy-storage solutions for transportation systems, consumer electronics, renewable energy buffering, and next-generation wearable technologies [45].

3.8. Challenges and Limitations

Despite their rapidly growing popularity and promising electrochemical properties, spinel-based materials for supercapacitors still face several challenges and limitations that hinder their large-scale commercialization and real-world deployment, many of which arise from the inherent characteristics of transition-metal oxides and the complexities of device engineering. One of the primary challenges is the moderate intrinsic electronic conductivity of many spinels, which, although superior to single-metal oxides, remains lower than desired for high-power applications, especially when the material is used in thick electrode layers where electron pathways become longer and internal resistance increases, resulting in voltage drop, heat generation, and reduced rate capability. Another major limitation is ion transport within the electrode; while nanoscale spinels offer short diffusion distances, large or agglomerated particles can trap active sites within the crystal bulk, making them difficult for electrolyte ions to reach, thereby reducing capacity utilization. The electrochemical reactions in spinels also involve structural changes associated with ion insertion/extraction and redox transitions, and although the 3D spinel framework is generally robust, repeated volume fluctuation can cause microcracks, mechanical degradation, and loss of electrical contact over long-term cycling, particularly in high-capacity systems where multiple oxidation states are accessed. Additionally, achieving controlled morphology, particle size distribution, and defect concentration during synthesis can be challenging, and small variations in hydrothermal conditions, calcination temperature, or precursor composition can lead to major differences in performance, making reproducibility a limiting factor for industrial manufacturing. The cost and environmental impact of some spinel compositions also raise concerns; elements such as cobalt are relatively expensive and associated with supply-chain and sustainability issues, while complex multistep syntheses require energy and chemical use that can increase environmental burden. From a practical device perspective, many laboratory tests rely on three-electrode configurations that may overestimate performance compared to real two-electrode systems, leading to gaps between academic reporting and commercial reality. Scaling up electrode coatings for large-area production introduces additional limitations, such as maintaining strong adhesion between active material and current collector, ensuring uniform thickness, and preventing cracking or delamination during drying or cycling. Electrolyte compatibility is also critical, as aqueous systems are safe and conductive but limited in voltage window, while organic or ionic liquid electrolytes offer higher energy density but bring increased cost, stricter sealing requirements, and potential flammability. Another significant limitation is the difficulty in balancing energy and power performance within a single material; spinels can provide high power density due to fast redox reactions, but their energy density still lags behind some battery materials, meaning hybrid cell designs are often required to achieve competitive performance. Finally, the long-term reliability and thermal stability of spinel-based devices under real operating conditions-such as temperature swings, continuous charge/discharge use, or prolonged high-voltage operation-remain insufficiently tested, and further studies are needed to fully understand degradation mechanisms and improve lifetime. Addressing these challenges will require advances in synthesis control, nanostructure engineering, cost reduction, scalable processing, and realistic device testing to unlock the full commercial potential of spinel-based supercapacitors [39,40].

3.9. Future Outlook and Perspectives

Looking ahead, the future of spinel-based supercapacitors is extremely promising, with research trends increasingly focused on designing materials and devices that bridge the gap between high-power supercapacitors and high-energy batteries [40], aiming to meet the growing demands of electric vehicles, grid-level renewable energy storage, Internet-of-Things systems, and next-generation portable electronics. Continued advances in material engineering are expected to center on improving conductivity, increasing ion-accessible surface area, and enhancing long-term structural stability, with strategies such as controlled defect introduction, surface functionalization, multi-ion doping, and crystal facet engineering poised to significantly improve redox kinetics and electron transport. Hybridization and compositing will remain crucial, especially as researchers combine spinels with carbon nanomaterials, MXenes, conductive polymers, or other metal oxides to create synergistic charge-storage mechanisms that simultaneously enhance energy density and power delivery while suppressing structural degradation during thousands of cycles. At the same time, progress in understanding charge-storage mechanisms through operando characterization methods such as in situ XRD, XPS, Raman spectroscopy, TEM analysis, and electrochemical impedance tracking will offer valuable insight into ion migration pathways, lattice evolution, and charge-transfer dynamics, enabling predictive modeling and rational material design rather than trial-and-error development. Machine learning and computational simulation are expected to play a larger role as well [27], accelerating the discovery of novel spinel compositions with optimized electronic structures, reduced activation energies, and higher redox-site density, potentially unlocking new families of high-performance materials beyond the commonly studied $NiCo_2O_4$, $MnCo_2O_4$, and $ZnCo_2O_4$ systems. From a device engineering standpoint, future research will focus on scalable processing methods such as spray deposition, roll-to-roll coating, 3D printing, electrodeposition, and ink formulation that can translate laboratory electrodes into industrial manufacturing while maintaining uniformity, mechanical robustness, and electrochemical efficiency. Flexible and wearable supercapacitors represent another major direction, where ultrathin spinel films, textile-supported electrodes, stretchable polymer electrolytes, and integrated smart sensors may enable high-performance power sources for health monitoring, soft robotics, and smart clothing. Environmentally friendly systems will also gain importance, with efforts aimed at reducing the use of expensive or supply-critical metals like cobalt, developing aqueous or gel-based electrolytes with larger safe voltage windows, and improving recycling and green synthesis routes to reduce the environmental footprint. In real-world applications, improving volumetric energy density and safety will be key to making spinel-based devices competitive with Li-ion batteries, and asymmetric or hybrid capacitor configurations will likely dominate because they allow higher operating voltages and balanced performance. Finally, long-term reliability studies under realistic conditions-high temperature, mechanical stress, continuous cycling, and large current loads-will be necessary to guarantee that spinel-based devices can operate reliably in commercial systems such as electric buses, regenerative braking, solar-grid buffering, emergency backup systems, and compact consumer electronics. By combining innovation in chemistry, nanostructure design, advanced characterization, scalable manufacturing, and realistic device integration, spinel-based supercapacitors are expected to evolve into mature, commercial energy-storage solutions that offer fast charging, high power, long life, environmental sustainability, and cost-effective performance for the next generation of global energy applications.

4. Conclusion

In conclusion, spinel-based materials have rapidly emerged as one of the most promising classes of electrode materials for next-generation supercapacitors [35], offering a unique combination of high theoretical capacitance, structural robustness, rich redox chemistry, and broad compositional tunability that allows researchers to optimize performance for diverse energy-storage applications. Their AB_2O_4 crystal structure provides multiple redox-active transition-metal sites, fast electron transport via mixed-valence conduction, and sufficient structural tolerance to withstand repeated ion insertion/extraction without catastrophic lattice collapse, enabling high cycling stability that is critical for long-term operation. However, achieving high practical performance depends heavily on controlling nanoscale morphology, composition, and interface characteristics, as these directly influence ion diffusion paths, electron mobility, and the degree to which active sites participate in charge storage. Significant progress has been made in engineering spinels into hollow structures, nanosheets, nanowires, hierarchical porous frameworks, and other architectures that maximize accessible surface area and accelerate reaction kinetics, while doping and multi-metal substitution have proven effective in enhancing conductivity, increasing redox-site density, and stabilizing reaction intermediates during cycling. The development of composite and hybrid systems-including combinations with carbon nanotubes, graphene, MXenes, conducting polymers, and other metal oxides-has helped overcome limitations such as moderate conductivity and structural stress, producing electrodes that deliver high power density, improved rate capability, and outstanding cycle life [32]. At the device level, spinel electrodes have demonstrated strong operation in both asymmetric and symmetric configurations, with real-world performance continuing to improve as researchers optimize electrode balancing, electrolyte selection, manufacturing methods, and current collector integration. Yet, challenges remain, including scalability of synthesis, environmental and supply-chain concerns for certain metals, performance gaps between laboratory three-electrode testing and real device operation, and the need for improved volumetric energy density to compete with established battery technologies. Future progress will require interdisciplinary advances in material design, operando characterization, theoretical modeling, large-scale fabrication, and device engineering to ensure that laboratory breakthroughs translate into reliable commercial systems. With ongoing innovations in green synthesis, flexible and wearable device integration, solid- and gel-electrolyte systems, and cost-reducing material substitutions, spinel-based supercapacitors are well positioned to play a major role in high-power energy-storage sectors, including electric mobility, renewable energy buffering, industrial power stabilization, next-generation IoT devices, and portable electronics. In summary, while challenges remain, the rapid pace of development, combined with the inherent advantages of spinel chemistry, suggests that these materials are moving steadily from laboratory research toward practical implementation, holding strong potential to contribute meaningfully to cleaner, more efficient, and more sustainable global energy solutions.

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Conflict of interest

The authors declare no conflict of interest.

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