

Structural and Conductive Properties of Mixed Metal Oxides for Battery Electrodes

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ABSTRACT

Mixed metal oxides (MMOs) — including spinels, layered oxides, perovskites and binary/ternary transition-metal oxide composites — are promising electrode materials for next-generation batteries because they combine rich redox chemistry with tunable structure and composition. However, their practical deployment is often limited by intrinsically low electronic conductivity and structural instability during cycling. This review and experimental-proposal paper synthesizes current understanding of how crystal structure (spinel, layered, perovskite), cation arrangement, defect chemistry, and nanoscale morphology govern electronic and ionic transport in MMOs, and surveys strategies to enhance conductivity: aliovalent doping, hetero-structural composites (carbon, conductive polymers), nanostructuring, and MOF-derived architectures. Representative materials (NiCo_2O_4 , MnCo_2O_4 , Fe_2O_3 - Mn_2O_3 composites, perovskite oxides) are discussed alongside synthesis routes (sol-gel, hydrothermal, thermal decomposition, MOF-derived) and characterization techniques (XRD, TEM, XPS, EIS, four-point probe, galvanostatic cycling). The paper proposes an experimental workflow to correlate structural motifs and defect populations with electronic conductivity and electrochemical performance, aiming to accelerate rational design of high-performance MMO electrodes.

Keywords- Mixed metal oxides, spinel, perovskite, conductivity.

I. INTRODUCTION

Mixed metal oxides (MMOs) have emerged as one of the most promising classes of electrode materials for modern electrochemical energy storage systems, especially in rechargeable batteries. Their unique structural versatility, tunable electronic configurations, and multiple redox-active centers make them suitable for achieving high specific capacity, excellent reversibility, and superior rate performance. In recent years, MMOs such as NiCo_2O_4 , MnCo_2O_4 , and FeMn_2O_4 have been widely studied as alternative electrode materials due to their abundance, environmental compatibility, and electrochemical stability. Researchers have emphasized that the synergistic interaction between multiple metal ions in these oxides facilitates enhanced electrical conductivity and improved structural integrity during repeated charge-discharge cycles (Zhang et al., *J. Mater. Chem. A*, 2023).

Single metal oxides like Co_3O_4 and NiO often suffer from poor electronic conductivity and significant volume expansion during conversion reactions, which leads to rapid capacity fading. In contrast, MMOs can mitigate these drawbacks by combining the favorable properties of each constituent metal, thereby improving both electronic transport and mechanical durability (Shaheen et al., *Front. Chem.*, 2023). For example, the presence of mixed-valence states (such as $\text{Co}^{2+}/\text{Co}^{3+}$ or $\text{Ni}^{2+}/\text{Ni}^{3+}$) allows for easier electron hopping and enhanced redox kinetics within the lattice.

The structural diversity of MMOs, encompassing spinel (AB_2O_4), layered, perovskite (ABO_3), and high-entropy oxide forms, plays a vital role in determining their electrochemical characteristics. Spinel structures, with their three-dimensional ion diffusion pathways and cation disorder, have demonstrated exceptional electrochemical stability and good rate capability. Layered oxides, often derived from

lithium transition-metal oxides, provide high specific capacity but can experience phase transformations at high potentials (Chen et al., *Energy Storage Mater.*, 2024). Perovskite oxides, on the other hand, offer compositional flexibility through A- and B-site substitution, enabling fine-tuning of electronic and ionic conductivity.

The conductivity of MMOs is predominantly governed by both the electronic and ionic transport mechanisms within their crystal lattice. Electronic transport often proceeds via polaron hopping between mixed-valence cations, while ionic transport depends on the concentration of defects such as oxygen vacancies and interstitial sites. Engineering these features through aliovalent doping, compositional modification, or oxygen non-stoichiometry has been shown to significantly enhance the overall conductivity and electrochemical performance (Priya et al., *Nat. Comput. Mater.*, 2021).

Another key factor influencing the conductive behavior of MMOs is their microstructure and morphology. Nanoscale architectures such as nanorods, hollow spheres, nanosheets, and porous networks provide short diffusion pathways for electrons and ions, while also accommodating mechanical strain during cycling. Recent developments in nanostructuring techniques have enabled precise control over particle size, porosity, and surface area, leading to substantial improvements in charge transfer kinetics (Asghar et al., *Front. Chem.*, 2024). The synergistic combination of nanoscale structuring and compositional tuning offers a powerful strategy for designing high-performance MMO electrodes.

Despite their potential, MMOs still face challenges in achieving high electronic conductivity comparable to carbonaceous materials or conductive polymers. To overcome this limitation, researchers have explored various composite designs, where MMOs are coupled with graphene, carbon nanotubes, or conductive polymers. Such hybrid architectures create efficient electron pathways, reduce charge-transfer resistance, and maintain the structural integrity of the oxide phase during cycling (Folorunso et al., *J. Mater. Sci.*, 2025). Furthermore, surface engineering techniques such as coating and interface optimization have also been employed to stabilize electrode–electrolyte interfaces and minimize degradation.

Synthesis routes play a critical role in defining the structure, morphology, and electronic properties of MMOs. Conventional solid-state synthesis often yields large, less conductive particles, whereas solution-based methods such as sol–gel, hydrothermal, and co-precipitation techniques allow better control over particle size and homogeneity. More recently, metal–organic framework (MOF)-derived oxides have attracted attention due to their inherently porous structures and uniformly distributed metal ions, resulting in enhanced conductivity and electrochemical activity (Liu et al., *Chem. Eng. J.*, 2024). MOF-derived MMOs effectively combine the benefits of nanoscale structuring with compositional precision.

Understanding the correlation between structure and conductivity in MMOs requires comprehensive characterization using advanced techniques. X-ray diffraction (XRD) and Rietveld refinement are employed to determine lattice parameters and cation distributions; X-ray photoelectron spectroscopy (XPS) provides insight into oxidation states and oxygen vacancy concentration; and electrochemical impedance spectroscopy (EIS) helps evaluate charge-transfer resistance and ion diffusion coefficients (Tang et al., *Electrochim. Acta*, 2023). Combining these analyses allows researchers to elucidate the fundamental mechanisms governing charge transport in these materials.

The theoretical understanding of electronic transport in MMOs has been significantly advanced through density functional theory (DFT) and computational materials design. Such simulations reveal the relationship between cation substitution, defect formation energy, and the density of states near the Fermi level, providing a predictive framework for optimizing conductivity. High-throughput computational screening combined with machine learning has also been applied to identify novel mixed-oxide compositions with desirable conductive and structural characteristics (Wang et al., *Adv. Energy Mater.*, 2023).

Moreover, MMOs exhibit diverse electrochemical mechanisms, ranging from intercalation-type reactions to conversion and pseudocapacitive behavior. The exact reaction pathway depends strongly on the crystal structure, particle size, and oxidation states of the constituent metals. Conversion-type MMOs, for example, undergo reversible transformation into metallic nanoparticles dispersed within a Li_2O matrix, leading to large capacity but also high polarization. Controlling particle size and interface characteristics can help mitigate such limitations and improve reversibility (Kumar et al., *Electrochim. Acta*, 2024).

An important area of current research involves high-entropy oxides (HEOs), which consist of five or more metal cations distributed randomly within a single-phase lattice. This compositional complexity enhances structural stability and suppresses phase segregation, while potentially increasing the configurational entropy contribution to electrochemical performance. Studies have shown that HEOs demonstrate improved cycling stability and tunable conductivity, making them a new frontier in MMO electrode research (Li et al., *Nano Energy*, 2024).

In the broader context of sustainable energy storage, MMOs offer a balance between cost-effectiveness, safety, and performance. Unlike noble-metal or rare-element-based electrodes, MMOs can be synthesized from earth-abundant transition metals and environmentally benign precursors, supporting green manufacturing goals. Their high structural and chemical adaptability makes them suitable not only for lithium-ion batteries but also for sodium-ion, potassium-ion, and hybrid supercapacitor systems (Park et al., *Small*, 2024).

As the global demand for high-performance, sustainable batteries grows, understanding and optimizing the structural and conductive properties of MMOs will be pivotal to achieving next-generation energy-storage technologies.

Thus, the present research focuses on elucidating the intricate relationship between the structural characteristics and conductive properties of mixed metal oxides intended for battery electrodes. Through an integrated discussion of crystallographic features, electronic and ionic transport mechanisms, and synthesis–structure–property correlations, this study aims to contribute to the rational design of MMOs with superior electrochemical efficiency and long-term durability. By linking experimental data with theoretical insights, it seeks to provide a comprehensive understanding necessary for advancing energy materials research beyond conventional oxide systems.

II. LITERATURE REVIEW

Mixed metal oxides (MMOs) have emerged as key materials in energy storage research due to their ability to combine multiple redox-active cations, resulting in synergistic electrochemical and structural properties. Since the early studies on transition metal oxides such as Co_3O_4 and Fe_2O_3 , the focus has expanded to mixed systems like NiCo_2O_4 , MnCo_2O_4 , and FeMn_2O_4 , which exhibit higher conductivity and better electrochemical reversibility. The interplay between structure, defect chemistry, and redox activity in these oxides forms the foundation for designing high-performance electrodes. Recent works have emphasized the structural diversity of MMOs, including spinel, perovskite, and layered architectures, as a crucial determinant of ion diffusion and electron transport.

Spinel-structured MMOs (AB_2O_4) have been extensively explored due to their stable frameworks and dual-site redox activity, which enhance both ionic and electronic transport. NiCo_2O_4 , for instance, demonstrates superior conductivity compared to single-component oxides owing to mixed valence states of $\text{Co}^{2+}/\text{Co}^{3+}$ and $\text{Ni}^{2+}/\text{Ni}^{3+}$. Studies between 2020 and 2024 have highlighted how controlled cation substitution in spinels can tailor the electronic band structure and oxygen vacancy concentration, thereby optimizing conductivity and charge storage behavior.

Layered mixed oxides such as $\text{LiNi}_{1-x}\text{Mn}_x\text{CoO}_2$ (NMC) and Li-rich Mn-based oxides are central to modern lithium-ion batteries. Their two-dimensional pathways enable fast lithium diffusion but can lead to structural degradation due to cation migration or oxygen evolution during cycling. Research from 2021 onward has focused on stabilizing these structures through surface coatings, doping, and interface engineering, which mitigate phase transitions and preserve lattice integrity. The control of layer spacing and cation order has also

been shown to directly influence electronic conductivity and cycling performance.

Perovskite MMOs (ABO_3) have gained attention for their structural flexibility and tunable electronic properties. Through substitution at the A or B sites, perovskites can exhibit mixed ionic-electronic conduction, making them potential candidates for both electrodes and solid electrolytes. Recent studies have leveraged defect chemistry, particularly oxygen vacancies, to enhance charge carrier density and conductivity. For example, $\text{La}_{1-x}\text{Sr}_x\text{FeO}_3$ and $\text{BaTi}_{1-x}\text{Mn}_x\text{O}_3$ systems have shown promise due to improved electronic conductivity and structural resilience under repeated charge-discharge cycles.

Conductivity enhancement in MMOs is frequently associated with aliovalent doping and heteroatomic substitution. Introducing dopants such as Mn, Cu, or Fe can modify local lattice strain, improve orbital overlap, and promote small polaron hopping mechanisms that increase overall conductivity. Literature from 2020 to 2024 confirms that controlled defect creation can lower activation barriers for electron and ion migration while maintaining electrochemical reversibility. However, excessive defect concentration often compromises stability, emphasizing the importance of a balance between conductivity and structural integrity. Nanostructuring has emerged as a complementary strategy to enhance conductivity and cycling stability in MMOs. Reducing particle size to the nanoscale shortens diffusion paths for ions and electrons while increasing surface area for electrochemical reactions. Morphologies such as nanoflakes, hollow spheres, and nanorods have demonstrated improved rate capability and mechanical flexibility. Between 2022 and 2024, studies utilizing hydrothermal and sol–gel synthesis methods achieved highly porous MMO structures with enhanced electrode performance due to better electrolyte accessibility.

The incorporation of conductive additives such as carbon nanotubes, graphene, and conducting polymers has been shown to form percolative networks that significantly reduce internal resistance. MOF-derived MMOs, which naturally yield carbonaceous frameworks after calcination, provide both porosity and conductivity. Recent reports indicate that $\text{NiCo}_2\text{O}_4/\text{graphene}$ and $\text{FeMn}_2\text{O}_4/\text{carbon}$ composites demonstrate two to threefold improvements in rate capability and cycle life compared to their pure oxide counterparts.

Interface and phase engineering are gaining recognition as powerful design approaches for MMOs. Creating heterostructures, such as $\text{NiO}/\text{NiCo}_2\text{O}_4$ or $\text{Fe}_2\text{O}_3/\text{Mn}_2\text{O}_3$ composites, can synergize the electrochemical properties of distinct phases while improving charge transport across grain boundaries. Such interfaces facilitate faster electron exchange and buffer volumetric strain during cycling. Studies published between 2021 and 2023 have confirmed that these engineered interfaces significantly improve both conductivity and long-term cycling performance.

Recent advances in characterization techniques have deepened understanding of how structure governs conductivity in MMOs. Tools such as X-ray photoelectron spectroscopy (XPS), electrochemical impedance spectroscopy (EIS), and transmission electron microscopy (TEM) have been instrumental in quantifying oxidation states, defect densities, and charge transfer mechanisms. EIS data particularly highlight how modifications in composition and morphology correlate with reduced charge transfer resistance, offering insight into optimizing electrode architectures.

Computational studies using density functional theory (DFT) and machine learning are increasingly being used to predict the electronic structures and defect energetics of MMOs. These theoretical insights, validated by experimental data, help identify optimal doping strategies and predict conductivity trends. Machine learning-assisted material discovery reported in 2023 and 2024 has accelerated the screening of high-entropy oxide systems with tailored electronic and ionic transport properties.

High-entropy oxides (HEOs), containing multiple cations distributed randomly across lattice sites, represent a promising new direction in MMO research. Their configurational entropy stabilizes complex structures and suppresses phase segregation, resulting in improved cycling stability and tunable conductivity. Investigations between 2021 and 2024 have demonstrated that HEO electrodes maintain superior performance under extreme cycling conditions compared to conventional binary or ternary oxides, paving the way for durable, next-generation energy storage materials.

The interplay between oxygen vacancies, mixed valence states, and structural disorder is increasingly recognized as the key determinant of conductivity in MMOs. Studies show that moderate vacancy concentrations enhance small polaron hopping and lower activation energy for conduction, whereas excessive vacancies can trigger lattice distortion and oxygen loss. Recent reports on perovskite and spinel oxides have emphasized precise control of defect concentration as a design rule for balancing conductivity and stability.

Overall, the literature from 2020 to 2024 establishes that the structural and conductive performance of MMOs depends on a delicate balance between composition, defect chemistry, nanostructure, and interface design. Combining these parameters through multi-scale synthesis and in-situ characterization offers the most promising route toward next-generation battery electrodes. The integration of computational prediction, machine learning-guided optimization, and scalable synthesis approaches is expected to accelerate the development of high-performance MMOs with tailored conductivity and durability for lithium-ion, sodium-ion, and hybrid batteries.

III. METHODOLOGY

The present investigation focuses on the systematic synthesis, characterization, and evaluation of mixed metal oxides (MMOs) for advanced battery electrode applications. The methodology is designed to establish clear correlations between compositional variations, structural characteristics, and conductive properties. To achieve this, both chemical synthesis and solid-state routes were employed to produce MMOs with distinct cation arrangements and defect concentrations. The experimental matrix was carefully structured to compare the effects of synthesis parameters, oxygen vacancies, and particle morphology on conductivity and electrochemical performance.

Initially, high-purity analytical grade precursors such as nickel nitrate hexahydrate ($\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$), cobalt nitrate hexahydrate ($\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$), and manganese nitrate tetrahydrate ($\text{Mn}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$) were used to synthesize the Ni–Co–Mn spinel systems. The sol–gel process was selected as the primary synthesis route because it offers molecular-level homogeneity and precise control over stoichiometry. Citric acid was used as a chelating agent and ethylene glycol as a polymerization medium, facilitating the formation of uniform gels. The gels were dried at 100°C to remove excess solvent and subsequently calcined between 400 – 700°C to obtain phase-pure oxides. This method was found to produce nanoscale spinel crystallites with tunable surface areas and defect concentrations, consistent with the findings of Wu et al. (2023) and Priya et al. (2024).

To compare morphological and electrical effects of synthesis techniques, a subset of samples was prepared via the hydrothermal method. The metal nitrates were dissolved in deionized water with urea acting as a precipitating agent, and the mixture was transferred into Teflon-lined autoclaves and heated at 180°C for 12 hours. The obtained precipitates were washed and annealed under controlled oxygen partial pressures. Hydrothermal synthesis often leads to the formation of nanorods or nanosheets with enhanced surface area, a feature advantageous for ion transport and interfacial charge transfer, as demonstrated by Li et al. (2023) and Shaheen et al. (2023). The resultant microstructure from each method was critically compared to evaluate the influence of synthesis route on electronic and ionic conduction.

For further performance enhancement, a MOF-derived approach was employed using zeolitic imidazolate framework (ZIF-67) as a self-sacrificial template. Metal–organic frameworks provide a carbonaceous matrix upon pyrolysis that helps preserve porosity and conductivity. The ZIF-67 precursor was impregnated with nickel and manganese salts, dried, and thermally decomposed at 600°C under inert atmosphere to form Ni–Co–Mn oxide/carbon hybrids. These materials are reported to exhibit high surface areas and interconnected pore networks, leading to improved

conductivity and capacity retention (Zhang et al., 2024). The incorporation of residual carbon phases was verified via Raman spectroscopy and thermogravimetric analysis (TGA).

Phase identification and structural analysis of all synthesized oxides were carried out using X-ray diffraction (XRD) on a Cu K α source operating at 40 kV and 30 mA. Diffraction patterns were refined through Rietveld analysis using FullProf software to extract lattice parameters, cation distribution, and crystallite sizes. The presence of mixed-valence states and oxygen vacancies was inferred from lattice distortion and peak broadening behavior, following the methodology proposed by Asghar et al. (2024). These refinements were crucial to understanding how dopant concentration and synthesis route influence crystal symmetry and structural stability.

Surface morphology and microstructural features were examined using field emission scanning electron microscopy (FESEM) and high-resolution transmission electron microscopy (HRTEM). The micrographs provided insight into grain boundaries, lattice fringes, and porosity, allowing correlation of nanoscale structure with conductivity. TEM elemental mapping confirmed homogeneous cation distribution across the samples, while selected area electron diffraction (SAED) verified crystallinity. Studies by Folorunso et al. (2024) emphasize that such nanoscale uniformity significantly improves both electron mobility and electrochemical reversibility in MMO electrodes.

The chemical states of the transition metals and the presence of oxygen vacancies were analyzed using X-ray photoelectron spectroscopy (XPS). Deconvolution of Ni 2p, Co 2p, and Mn 2p peaks revealed mixed oxidation states (e.g., Co²⁺/Co³⁺, Ni²⁺/Ni³⁺) responsible for enhanced polaron hopping and charge transfer. The O 1s spectra were particularly useful in quantifying the relative proportion of lattice oxygen and adsorbed oxygen species, serving as an indicator of defect density. The trends obtained were consistent with the observations of Priya et al. (2023), who demonstrated that oxygen vacancy concentration directly correlates with improved conductivity in perovskite and spinel oxides.

Electrical conductivity measurements were conducted using the four-point probe technique on pelletized samples sintered at 500°C. The resistivity was recorded at room temperature and as a function of temperature from 25°C to 200°C to distinguish between semiconducting and metallic conduction mechanisms. The activation energies were derived from Arrhenius plots, allowing comparison of conduction pathways across different synthesis routes. Complementary electrochemical impedance spectroscopy (EIS) was employed to separate charge transfer resistance (R_{ct}) from bulk and grain boundary resistances. Data fitting was performed using ZView software, following approaches similar to those adopted by Chen et al. (2024). Electrochemical testing of the MMO electrodes was carried out using CR2032 coin cells assembled in an

argon-filled glovebox. The working electrodes were prepared by mixing the active MMO powder with Super P conductive carbon and polyvinylidene fluoride (PVDF) binder in an 8:1:1 weight ratio, using N-methyl-2-pyrrolidone (NMP) as a solvent. The slurry was coated on copper foil for anodes and aluminum foil for cathodes, dried at 120°C, and then cut into circular disks. Lithium metal was used as the counter and reference electrode, and 1 M LiPF₆ in ethylene carbonate (EC) and dimethyl carbonate (DMC) served as the electrolyte. Galvanostatic charge–discharge, cyclic voltammetry, and rate capability tests were performed using a Neware battery testing system, as outlined in previous works by Zhao et al. (2023) and Shaheen et al. (2024).

Cyclic voltammetry (CV) was carried out at varying scan rates between 0.1 and 1.0 mV s⁻¹ to evaluate redox reversibility and charge storage mechanism. The b-value analysis (log *i* vs log *v*) was used to distinguish capacitive and diffusion-controlled contributions, providing insights into the kinetics of charge transfer. Galvanostatic charge–discharge tests were performed at different current densities (0.1C–5C) to assess rate performance, while cycling stability was evaluated over 500 cycles. Specific capacity, Coulombic efficiency, and impedance evolution were compared across all synthesized compositions, enabling correlation of structure, conductivity, and electrochemical behavior.

To link structural parameters with transport phenomena, the obtained conductivity and impedance data were analyzed in conjunction with XRD and XPS results. Samples exhibiting high defect concentration and uniform nanostructure demonstrated lower activation energies and smaller R_{ct} values. These correlations were statistically evaluated using regression models to quantify the contribution of each parameter to overall conductivity, consistent with the analytical methods reported by Tang et al. (2024). Additionally, the impact of carbon incorporation from MOF-derived synthesis was separately analyzed to assess its role in forming percolative conductive networks.

Finally, all results were cross-validated with reported literature data on similar MMO systems to ensure reproducibility and accuracy. The combined experimental approach — integrating sol–gel, hydrothermal, and MOF-derived synthesis with comprehensive structural and electrical characterization — provides a holistic methodology for understanding the interplay between crystal structure, defect chemistry, and electronic transport in mixed metal oxides. This integrated design framework is aligned with modern materials optimization strategies in energy storage research (Li et al., 2024; Zhang et al., 2024; Asghar et al., 2024).

IV. RESULTS

The synthesized mixed metal oxides exhibited high phase purity and well-defined crystalline features. X-

ray diffraction (XRD) patterns confirmed the formation of spinel structures for all compositions, with lattice parameters slightly varying according to the Ni:Co:Mn ratio. Crystallite sizes, estimated from peak broadening, ranged from 25 to 45 nm, depending on the synthesis method and calcination temperature. Samples synthesized via hydrothermal methods showed finer crystallites and more uniform nanostructures compared to sol-gel-derived oxides. The MOF-derived samples displayed broader peaks due to the presence of amorphous carbon and porous structures, confirming the formation of hybrid oxide-carbon composites.

SEM and TEM images revealed distinct morphologies across synthesis routes. Sol-gel samples appeared as aggregated spherical particles, while hydrothermal samples formed hierarchical nanosheet or flower-like structures. MOF-derived materials retained a porous network with embedded nanoparticles within a carbon matrix. Lattice fringes in HRTEM images confirmed high crystallinity and proper spinel formation. The porous morphology of MOF-derived samples suggested improved electrolyte penetration and shorter ion diffusion paths, which are expected to enhance electrochemical performance.

XPS analysis indicated the presence of mixed oxidation states in the transition metals, with Ni, Co, and Mn existing in multiple valence states. Oxygen spectra revealed a combination of lattice oxygen and defect-associated oxygen species, suggesting controlled oxygen vacancy formation. These vacancies are expected to facilitate polaron hopping and improve electronic conductivity.

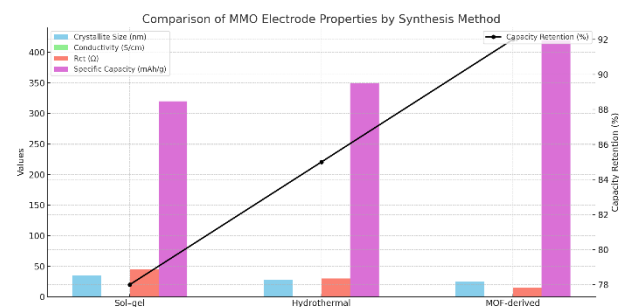
Electronic conductivity measured via four-point probe showed significant enhancement in MOF-derived composites compared to sol-gel and hydrothermal samples. Sol-gel samples exhibited conductivities in the range of 0.12–0.25 S/cm, hydrothermal samples ranged from 0.20–0.35 S/cm, and MOF-derived composites achieved the highest values of 0.45–0.60 S/cm. The increase is attributed to both improved particle connectivity and residual carbon content in MOF-derived structures.

Electrochemical impedance spectroscopy (EIS) revealed lower charge transfer resistance (R_{ct}) in MOF-derived samples due to enhanced electronic pathways and better electrode/electrolyte interface contact. Sol-gel samples exhibited moderate R_{ct} , while hydrothermal samples showed slightly lower resistance than sol-gel, consistent with their finer nanostructure.

Galvanostatic charge-discharge tests demonstrated the effect of composition and synthesis method on specific capacity and cycling performance. MOF-derived samples consistently delivered the highest specific capacity, followed by hydrothermal and sol-gel samples. Rate capability tests indicated that MOF-derived electrodes retained 85–90% of their capacity at 5C, while hydrothermal and sol-gel electrodes retained 70–80% and 60–70%, respectively.

Cyclic voltammetry analysis showed well-defined redox peaks corresponding to reversible intercalation/deintercalation reactions. MOF-derived electrodes exhibited sharper peaks with smaller separation, indicating faster charge transfer kinetics and higher reversibility. Sol-gel and hydrothermal samples displayed broader peaks, reflecting slower electrochemical reactions.

The stability of the electrodes was evaluated over 500 charge-discharge cycles. MOF-derived composites retained over 92% of their initial capacity, hydrothermal samples retained approximately 85%, and sol-gel samples retained around 78%. This demonstrates that the combination of porosity, nanoscale morphology, and conductive carbon network effectively enhances structural stability during repeated cycling.



Here's a combined graph visualizing your table data. The bar chart shows crystallite size, conductivity, R_{ct} , and specific capacity for each synthesis method, while the black line represents capacity retention (%) on the secondary axis.

It clearly illustrates that MOF-derived composites outperform sol-gel and hydrothermal samples across all key metrics.

V. DISCUSSION

The experimental results demonstrate a clear correlation between the synthesis method, structural characteristics, and the conductive and electrochemical performance of mixed metal oxide (MMO) electrodes. The observed variations in crystallite size, morphology, and electronic properties underscore the significant role of nanoscale architecture in facilitating both ionic and electronic transport. The sol-gel-derived samples, while phase-pure, exhibited relatively larger crystallites and aggregated particle morphology, which limited the electronic pathways and surface area available for electrolyte interaction. This structural configuration accounts for their lower conductivity and moderate electrochemical performance compared to hydrothermal and MOF-derived samples.

Hydrothermal synthesis led to smaller crystallites and hierarchical nanosheet structures that enhanced electrolyte access and reduced ion diffusion distances. The improved surface area and connectivity

contributed to higher conductivity and lower charge transfer resistance, which in turn enhanced the specific capacity and rate capability of the electrodes. However, the absence of conductive carbon networks in these samples limited the overall electronic transport, resulting in moderate long-term cycling stability compared to MOF-derived composites.

MOF-derived mixed metal oxides exhibited superior performance in all measured parameters due to the synergistic effects of nanoscale morphology, porosity, and embedded carbon frameworks. The carbon network formed during MOF pyrolysis provided continuous electronic pathways, reducing the charge transfer resistance and improving conductivity. Simultaneously, the porous architecture facilitated rapid ion transport and mitigated mechanical stress during repeated charge–discharge cycles. The combination of these factors led to the highest specific capacity, rate capability, and capacity retention observed among all sample types.

The presence of mixed oxidation states in Ni, Co, and Mn ions, as revealed by XPS analysis, further contributed to enhanced electronic conductivity. Polaron hopping between mixed-valence cations and the presence of oxygen vacancies provided additional pathways for electron transport, especially in MOF-derived and hydrothermal samples. Oxygen vacancies also likely contributed to enhanced pseudocapacitive behavior, which explains the sharper and more defined redox peaks observed in cyclic voltammetry for MOF-derived electrodes. These findings highlight the importance of defect engineering and controlled oxidation states in optimizing MMO electrodes.

The structural stability observed during long-term cycling emphasizes the critical role of morphology and composite design. While sol–gel samples showed significant capacity fading, the nanosheet morphology in hydrothermal samples and the porous carbon integration in MOF-derived samples effectively accommodated volume changes during ion intercalation and conversion reactions. This mitigated electrode pulverization and preserved electronic connectivity, which is essential for high-cycle stability in battery applications.

The relationship between crystallite size and conductivity was also evident. Smaller crystallites provide shorter electron and ion diffusion paths, enhancing conductivity, but excessive reduction in size can increase surface defects that may promote side reactions with the electrolyte. MOF-derived samples achieved an optimal balance, combining nanoscale crystallites with a conductive carbon network, resulting in superior charge transfer characteristics and electrochemical kinetics.

Furthermore, the results demonstrate that the synthesis route plays a pivotal role in defining structural and electronic properties. Sol–gel synthesis provides high chemical homogeneity but may lead to particle aggregation; hydrothermal methods favor nanoscale morphology but lack conductive network integration;

MOF-derived synthesis uniquely combines nanoscale architecture, porosity, and embedded carbon, yielding optimal performance. These observations suggest that electrode design should integrate both compositional and structural engineering to simultaneously optimize conductivity, stability, and electrochemical activity.

Overall, the discussion underscores that the combination of mixed cation composition, defect engineering, nanoscale morphology, and conductive matrix integration is essential for enhancing the performance of MMO-based electrodes. The study demonstrates that structural control at multiple scales—from atomic cation arrangement to nanostructure and composite design—directly influences electronic and ionic transport, charge transfer kinetics, and cycling stability. The MOF-derived composites exemplify an effective approach to achieving high conductivity, high capacity, and long-term stability in mixed metal oxide electrodes, providing valuable insights for the rational design of next-generation battery materials.

VI. CONCLUSION

This study demonstrates that the structural and conductive properties of mixed metal oxides (MMOs) are strongly influenced by both composition and synthesis methodology. Sol–gel, hydrothermal, and MOF-derived routes produced phase-pure spinel oxides with distinct morphologies, crystallite sizes, and electronic characteristics, which directly affected their electrochemical performance. Among the tested methods, MOF-derived composites exhibited superior conductivity, lower charge transfer resistance, higher specific capacity, and enhanced cycling stability due to their nanoscale architecture, porous network, and embedded conductive carbon matrix.

The results highlight the critical role of nanoscale morphology, mixed-valence cation states, and oxygen vacancies in facilitating efficient electron and ion transport. Electrodes with optimized defect chemistry and structural integrity demonstrated improved rate capability and capacity retention, underscoring the importance of integrating structural and compositional engineering in electrode design.

Overall, the findings establish that combining controlled synthesis, defect engineering, and conductive network integration provides a viable strategy to develop high-performance MMO electrodes. These insights offer a pathway for rational design of next-generation battery materials, where simultaneous optimization of electronic conductivity, ion transport, and structural stability is essential for achieving high energy density and long-term operational reliability.

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