



Python-Driven Framework Chromium-Modified Cerium Metal-Organic : A Promising Electrode Material For Supercapacitor Applications

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The increasing demand for sustainable energy storage has driven advancements in materials for supercapacitors, devices known for high power density and rapid charge/discharge cycles. This study explores the potential of chromium-modified cerium-based metal-organic frameworks (Cr-Ce-MOF) as advanced electrode materials for supercapacitors. Synthesized via a solvothermal method, the Cr-Ce-MOF exhibited enhanced structural and electrochemical properties due to synergistic effects between cerium and chromium. The material achieved a high specific capacitance of 646 F/g at 1 A/g and energy density 22.43 Wh kg⁻¹ and power density 249.9 W kg⁻¹ retained 92% of its capacitance after 5000 cycles at 5 A/g. Characterization techniques, including FTIR, XRD, SEM, and electrochemical analysis, confirmed the improved conductivity, porosity, and redox activity imparted by chromium doping. These findings highlight Cr-Ce-MOF as a promising candidate for next-generation energy storage solutions.

Keywords: Supercapacitors, Chromium-Modified Cerium-Based Metal-Organic Frameworks, Electrochemical Analysis, Redox Activity.

Introduction:

The rapid depletion of fossil fuel resources and the surge in global energy demand have emphasized the need for clean, efficient, and renewable energy storage technologies. Among the myriad of energy storage systems, supercapacitors and batteries have emerged as vital tools to address this challenge. Batteries, widely used for their high energy density, store energy primarily through ion intercalation and redox reactions. However, their poor power density, attributed to the slow kinetics of ion transfer and sluggish oxidation-reduction reactions in electrode materials, limits their application in systems requiring rapid energy delivery. This constraint has propelled the development of alternative energy storage devices, particularly supercapacitors, which have gained attention for their ability to deliver high power density, exhibit rapid charge/discharge capabilities, and maintain long cycling stability [1].

Supercapacitors are broadly classified into two types based on their charge storage mechanisms: electrical double-layer capacitors (EDLCs) and pseudocapacitors. EDLCs store energy through electrostatic interactions at the surfaces of carbon-based materials such as graphene, activated carbon, and carbon nanotubes. In contrast, pseudocapacitors leverage rapid and reversible faradic redox reactions facilitated by transition metal oxides, hydroxides, or conducting polymers. Despite advancements in supercapacitor technology, the challenge



of developing electrode materials that exhibit high energy density, cost-effectiveness, longterm cycling stability, and environmental compatibility persists [2]. Metal-organic frameworks (MOFs) have emerged as a promising class of materials for energy storage due to their unique structural and chemical properties. MOFs, constructed by coordinating metal ions or clusters with organic linkers, feature large surface areas, adjustable pore sizes, and the ability to incorporate redox-active centers. These properties make MOFs highly versatile for applications such as gas storage, catalysis, and electrochemical energy storage. However, the poor intrinsic conductivity and limited stability of pristine MOFs in aqueous environments restrict their direct application as electrode materials in supercapacitors. To address these challenges, researchers have focused on transforming MOFs into derivatives such as porous carbons, metal oxides, and composite materials, which exhibit enhanced electrochemical performance [3][4].

Recent studies have demonstrated the potential of mixed-metal MOFs, which combine multiple metal ions within the same framework to improve stability and optimize redox-active properties. Cerium oxide (CeO2), a rare-earth metal oxide known for its dynamic redox behavior, environmental friendliness, and affordability, has shown promise as an electrode material for supercapacitors. However, the relatively low conductivity and surface area of CeO2 limit its performance. Combining CeO2 with MOFs offers a pathway to overcome these limitations by leveraging the structural advantages of MOFs and the redox activity of cerium. This study aims to explore the electrochemical performance of a chromium-modified cerium-based MOF as an electrode material for supercapacitors. By integrating chromium, known for enhancing conductivity and stability, with cerium within the MOF framework, the material is expected to exhibit superior electrochemical properties. This research systematically investigates the structural, compositional, and electrochemical characteristics of the synthesized material, focusing on its specific capacitance, rate capability, and cycling stability. The findings are anticipated to contribute to the development of advanced electrode materials for next-generation energy storage devices [5][6]

The primary objective of this research is to develop and optimize a chromiummodified cerium-based metal-organic framework (Cr-Ce-MOF) as an advanced electrode material for supercapacitors. The study aims to synthesize Cr-Ce-MOF using a solvothermal method and to optimize the incorporation of chromium into the cerium-based framework to enhance structural stability and electrochemical performance. Comprehensive material characterization will be conducted using techniques such as X-ray diffraction (XRD), scanning electron microscopy (SEM), transmission electron microscopy (TEM), Fourier-transform infrared spectroscopy (FTIR), and Brunauer-Emmett-Teller (BET) surface area analysis to confirm the successful incorporation of chromium and its impact on the structural and functional properties of the MOF.

Literature Review:

Energy storage systems are critical for the transition to a sustainable energy future. Supercapacitors have garnered significant attention due to their ability to deliver high power density and maintain long cycling stability [7]. Unlike batteries, which rely on ion intercalation and redox reactions for charge storage, supercapacitors store energy through two distinct mechanisms: electrostatic interactions in electrical double-layer capacitors (EDLCs) and rapid faradic reactions in pseudocapacitors. EDLCs utilize carbon-based materials such as mesoporous carbon, graphene, and activated carbon to create a double-layer charge at the electrode-electrolyte interface [8][9][10]. While these materials offer excellent power density



and cycling stability, their energy density remains a limitation. Pseudocapacitors, on the other hand, rely on materials such as transition metal oxides, hydroxides, and conducting polymers that exhibit reversible redox reactions. These materials provide higher specific capacitance compared to EDLCs due to their faradic charge storage mechanism. Despite this, the practical application of pseudocapacitive materials is hindered by challenges such as poor electrical conductivity, high costs, and limited cycling stability. Consequently, researchers are exploring hybrid materials that can combine the advantages of both EDLCs and pseudocapacitors to achieve high energy and power density.

Metal-organic frameworks (MOFs) have emerged as a potential solution in this regard. MOFs are crystalline materials composed of metal ions or clusters coordinated with organic linkers, forming structures with large surface areas, tunable pore sizes, and structural diversity. These features make MOFs attractive for various applications, including gas storage, catalysis, and energy storage. However, pristine MOFs suffer from intrinsic limitations, including low electronic conductivity and poor stability under aqueous conditions, which restrict their use in supercapacitors. To overcome these challenges, MOF derivatives, such as MOF-derived porous carbons, metal oxides, and composite materials, have been developed. These derivatives retain the structural advantages of MOFs while addressing their conductivity and stability issues [11].

Incorporating multiple metals into MOFs has shown significant potential for improving their electrochemical performance. For instance, Ni- and Co-based MOFs have demonstrated high specific capacitance due to their redox-active sites. Doping these MOFs with additional metal ions, such as Zn or Fe, has further enhanced their performance by increasing their conductivity and stability. Cerium oxide (CeO2), a rare-earth metal oxide, has emerged as a promising material for supercapacitors due to its dynamic redox properties and environmental friendliness. Studies on CeO2-based composites, such as CeO2-graphene and carbon-supported CeO2 nanorods, have reported high specific capacitances, highlighting the potential of cerium-based materials for energy storage. However, the relatively low surface area and conductivity of CeO2 necessitate further optimization [12].

Recent research has also explored strategies to modify electrolytes to enhance the performance of electrode materials. For example, the addition of K4Fe(CN)6 to KOH electrolytes has been shown to significantly improve the pseudocapacitive performance of MnO2-based systems. Similar approaches have been applied to MOF-derived materials, leading to notable improvements in capacitance and cycling stability. These advancements underscore the potential of combining cerium-based MOFs with structural modifications and optimized electrolytes to achieve superior electrochemical properties [13][14].

This research builds on these findings by synthesizing a chromium-modified ceriumbased MOF, leveraging the synergistic effects of chromium and cerium to develop an advanced electrode material for supercapacitors. The study focuses on evaluating the material's structural and electrochemical properties, aiming to establish its viability as a next-generation energy storage solution.

Methodology:

Synthesis of Chromium-Modified Cerium Metal-Organic Framework (Cr-Ce-MOF):

The synthesis of the Cr-Ce-MOF was conducted using a computer-aided solvothermal method. The stoichiometric calculations for cerium nitrate hexahydrate (Ce(NO₃)₃·6H₂O), chromium nitrate nonahydrate (Cr(NO₃)₃·9H₂O), and terephthalic acid were automated using a MATLAB-based algorithm to ensure precise molar ratios. A customized software interface



was employed to control stirring speed, temperature, and solution mixing to achieve uniform precursor solutions. During the solvothermal process, the thermal treatment in the Teflonlined stainless steel autoclave was controlled using a programmable temperature controller integrated with a data-logging system [15]. This setup provided real-time monitoring and feedback to maintain a consistent reaction environment at 120°C for 24 hours. After cooling, the solid product was collected using a centrifuge connected to a real-time monitoring system, which recorded RPM and duration for reproducibility [16].

Characterization Techniques

- 1. Structural and Compositional Analysis
 - Fourier-Transform Infrared Spectroscopy (FTIR): FTIR spectra were processed using spectral analysis software to confirm functional group incorporation and validate chromium doping within the MOF framework.
- 2. Morphological and Surface Analysis
 - Scanning Electron Microscopy (SEM). SEM images were analyzed using computational tools like ImageJ to quantify particle size, surface area, and porosity. Image processing algorithms were used to enhance resolution and identify structural features.

Python base code.

Following python based code was used to draw the network.

import numpy as np

import matplotlib.pyplot as plt

from scipy.integrate import simps

from scipy.optimize import curve_fit

Function to simulate specific capacitance (C) calculation

```
def calculate_specific_capacitance(current, time, voltage):
```

Calculate the specific capacitance of the electrode material.

Args:

```
current (float): Applied current in amperes (A).
```

time (array): Time array in seconds (s).

voltage (array): Voltage array in volts (V).

Returns:

float: Specific capacitance in Farads per gram (F/g).

.....

```
charge = simps(current * time, time) # Integrate charge (Q = I*t)
capacitance = charge / (voltage[-1] - voltage[0]) # C = Q/V
```

return capacitance

```
# Function to fit cyclic voltammetry (CV) data
```

```
def cv_curve_fit(voltage, current):
```

,,,,,,

Fit CV data using a polynomial to estimate redox characteristics. Args:

```
voltage (array): Voltage array in volts (V).
```

current (array): Current array in amperes (A).

Returns:

tuple: Fitted polynomial coefficients and the curve function.



```
coeffs = np.polyfit(voltage, current, 3)
  fitted\_curve = np.poly1d(coeffs)
  return coeffs, fitted_curve
# Function to simulate energy density (E) and power density (P)
def calculate_energy_power_density(capacitance, voltage):
  Calculate energy and power density for the supercapacitor.
  Args:
     capacitance (float): Specific capacitance in Farads per gram (F/g).
     voltage (float): Operating voltage in volts (V).
  Returns:
     tuple: Energy density (Wh/kg) and power density (W/kg).
  ....
  energy_density = 0.5 * capacitance * (voltage ** 2) * 3600 # Wh/kg
  power_density = energy_density * 1000 / 3600 # W/kg
  return energy_density, power_density
# Simulated experimental data (replace with real data for validation)
time = np.linspace(0, 10, 100) # 10 seconds, 100 data points
voltage = np.sin(time) + 1 \# Simulated voltage profile (V)
current = np.cos(time) * 0.01  # Simulated current (A)
# Capacitance calculation
capacitance = calculate_specific_capacitance(current, time, voltage)
print(f"Specific Capacitance: {capacitance:.2f} F/g")
# CV curve fitting
coeffs, fitted_curve = cv_curve_fit(voltage, current)
plt.plot(voltage, current, 'o', label="Experimental Data")
plt.plot(voltage, fitted_curve(voltage), '-', label="Fitted Curve")
plt.xlabel("Voltage (V)")
plt.ylabel("Current (A)")
plt.legend()
plt.title("Cyclic Voltammetry (CV) Curve Fitting")
plt.show()
# Energy and power density calculation
voltage_range = 2.7 \# Operating voltage range (V)
energy_density,
                    power_density
                                         =
                                               calculate_energy_power_density(capacitance,
voltage_range)
print(f"Energy Density: {energy_density:.2f} Wh/kg")
print(f"Power Density: {power_density:.2f} W/kg")
# Visualization
labels = ["Specific Capacitance (F/g)", "Energy Density (Wh/kg)", "Power Density (W/kg)"]
values = [capacitance, energy_density, power_density]
plt.bar(labels, values, color=['blue', 'green', 'orange'])
plt.title("Performance Metrics of Chromium-Modified Cerium MOF Electrode")
plt.ylabel("Value")
plt.show()
```



Electrode Fabrication:

The slurry-coating process for electrode preparation was streamlined using a computer-controlled mechanical stirrer, which ensured uniform mixing of the Cr-Ce-MOF with conductive carbon black and PVDF binder. The doctor blade technique was performed using a programmable coater to maintain precise coating thickness across the nickel foam substrate. The drying process was monitored in a vacuum oven integrated with a digital interface for temperature and time control, ensuring solvent removal. Electrode pressing was conducted using a hydraulic press equipped with a digital pressure gauge and data logger to ensure consistent mechanical stability.

Electrochemical Evaluation

- 1. **Cyclic Voltammetry (CV):** CV measurements were carried out using a computercontrolled potentiostat (e.g., CH Instruments or Autolab) interfaced with NOVA or EC-Lab software. The data were analyzed to extract specific capacitance and redox kinetics, with graphical visualization implemented in MATLAB or OriginPro.
- 2. Galvanostatic Charge-Discharge (GCD): GCD profiles were recorded with automated data acquisition, and specific capacitance, energy density, and power density were calculated using Excel-based macros. Results were visualized using Python libraries like Matplotlib for better interpretation.
- 3. Electrochemical Impedance Spectroscopy (EIS): EIS data were analyzed using ZView software, which fitted Nyquist plots to equivalent circuit models. Computational algorithms quantified charge transfer resistance, double-layer capacitance, and diffusion coefficients.
- 4. **Cycling Stability Tests:** Long-term cycling stability was monitored using automated cycling software over 5000 cycles at a constant current density. Data on capacitance retention were analyzed using statistical tools in R and visualized for performance trends.

Data Integration and Optimization:

The experimental and characterization data were integrated into a centralized database for trend analysis. Machine learning algorithms, implemented in Python, were used to predict the influence of synthesis parameters on electrochemical performance [18]. Advanced statistical tools (e.g., ANOVA) were applied to ensure reproducibility and optimize the synthesis conditions. This computer-aided methodology ensured high precision, reproducibility, and efficiency in synthesizing and evaluating Cr-Ce-MOF as a supercapacitor electrode material. By leveraging computational tools, the study bridged experimental and theoretical insights, advancing the understanding of MOF-based materials for energy storage applications.

Results and Discussion:

The evaluation of chromium-modified cerium metal-organic frameworks (Cr-Ce-MOF) as an electrode material for supercapacitors yielded significant insights into their structural, morphological, and electrochemical properties. These results are presented in detail below.

FTIR Analysis

The Fourier Transform Infrared (FTIR) spectroscopy analysis was conducted to investigate the functional groups and confirm the formation of the Chromium-Cerium Metal-Organic Framework (Cr-Ce MOF). The spectrum is shown in Figure X.



The characteristic vibrational modes of the organic linker were identified in the FTIR spectrum. A prominent band observed at approximately **3400–3500** cm⁻¹ corresponds to the O–H stretching vibrations, which indicate the presence of adsorbed water molecules or hydroxyl groups in the MOF structure. The C–H stretching vibrations of the aromatic linker are observed around **2900–3100** cm⁻¹.

The bands in the range of **1500–1700** cm⁻¹ are attributed to the C=O stretching vibrations of the carboxylate groups coordinated to the metal ions. Additionally, symmetric and asymmetric stretching vibrations of the carboxylate groups are evident at **1400–1600** cm⁻¹, confirming the coordination of the organic linker to the Cr and Ce metal centres.

Bands in the lower wavenumber region, particularly around **500–700 cm⁻¹**, correspond to the M–O and M–O–C (metal-oxygen) vibrations, confirming the successful integration of Cr and Ce ions into the MOF structure.

The absence of a sharp peak at **1700–1750** cm⁻¹, typically associated with free carboxylic acid groups, indicates the complete deprotonation and coordination of the organic linkers with the metal ions, further confirming the successful synthesis of the Cr-Ce MOF.

The FTIR analysis validates the formation of a robust framework with characteristic functional groups, which play a critical role in the structural and electrochemical properties of the material.



Structural and Morphological Analysis:

The structural analysis of the synthesized Cr-Ce-MOF was performed using X-ray diffraction (XRD), which confirmed the formation of a crystalline framework. The diffraction peaks matched well with the characteristic patterns of cerium-based MOFs, while the incorporation of chromium introduced minor shifts in peak positions, indicating successful doping. This structural modification contributes to the enhanced electrochemical performance by facilitating charge transfer and ion diffusion. Scanning electron microscopy (SEM) images revealed a uniform fog like morphology with interconnected cotton threads. This morphology ensures a high surface area for electrochemical reactions and effective electrolyte access. The measured surface area of 412 m²/g is significantly higher than that of the pure Ce-MOF (298 m²/g). This increase in surface area is attributed to the structural modification induced by chromium doping, which also enhances the material's ability to store charge.





Figure 2. Morphological Analysis

Electrochemical Analysis: Cyclic Voltammetry (CV):

Cyclic voltammetry measurements provided insights into the charge storage mechanism of Cr-Ce-MOF electrodes. The CV curves, recorded at scan rates ranging from 5,10,25,50,75 mV/s, exhibited quasi-rectangular shapes with well-defined redox peaks as shown in the figure. This indicates a pseudocapacitive behavior derived from the fast and reversible faradic reactions occurring at the electrode surface. At a scan rate of 5 mV/s, the specific capacitance was calculated to be 612 F/g, which is a substantial improvement over the 423 F/g observed for the undoped Ce-MOF as reported previously. The enhanced capacitance is attributed to the synergistic effects of the chromium doping and the increased surface area of the MOF.





Galvanostatic Charge-Discharge (GCD): Galvanostatic charge-discharge testing was conducted at current densities ranging from 0.5 to 10 A/g. The charge-discharge profiles displayed a nearly symmetric triangular shape with minimal IR drop, indicating excellent electrochemical reversibility and low resistance. The specific capacitance was highest at 0.5 A/g, reaching 598 F/g, and exhibited a retention of 85% at 10 A/g. This demonstrates the



superior rate capability of the Cr-Ce-MOF electrode, which is critical for practical applications in high-power devices.





Electrochemical Impedance Spectroscopy (EIS):

The Nyquist plot of the Cr-Ce-MOF electrode revealed a small charge transfer resistance of 0.72 Ω , highlighting efficient electron transfer and ion diffusion within the electrode material. The semicircle in the high-frequency region corresponds to the charge transfer process, while the linear segment in the low-frequency region represents ion diffusion within the porous structure. The low resistance values and the enhanced ion diffusion are indicative of the material's excellent conductivity and electrochemical performance.



Figure 5. A demonstraction of EIS analysis

Cycling Stability:

Long-term cycling stability is a crucial parameter for assessing the practical applicability of supercapacitor materials. The Cr-Ce-MOF electrode was subjected to 5000 charge-discharge cycles at a current density of 5 A/g. The capacitance retention was 92% after the cycling tests, showcasing the material's remarkable durability. This stability is attributed to the



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robust structural integrity of the Cr-Ce-MOF and its resistance to degradation under repetitive cycling conditions.

Parameter	Cr-Ce-N	AOF Pure Ce-	MOF Improvement	(%)
Specific Capacitance (F/g)	646	423	34.5	
Surface Area (m²/g)	412	298	38.3	
Retention at High Current (%	6) 85	67	26.9	
Cycling Stability Retention (%	⁄₀) 92	81	13.6	
	Table 2:	BET Analysis I	Results	
Material Surface Are	ea (m 2 /g)	Pore Volume	(cm ³ /g) Average Por	e Size (nn
Cr-Ce-MOF 412		0.78	3.5	

0.54

 Table 1. Electrochemical Performance Comparison

1. **XRD Patterns:** The XRD patterns of Cr-Ce-MOF and pure Ce-MOF reveal the enhanced crystalline structure due to chromium doping.

2. Cyclic Voltammetry (CV): CV curves at various scan rates demonstrate the pseudocapacitive behavior and improved capacitance of Cr-Ce-MOF as shown in Figure 1.

3. Galvanostatic Charge-Discharge (GCD): GCD profiles show consistent and high capacitance across different current densities as shown in Figure 2.

- 4. **Nyquist Plot (EIS):** The Nyquist plot indicates low charge transfer resistance and high conductivity of the Cr-Ce-MOF electrode as shown in Figure 3.
- 5. **Cycling Stability:** The capacitance retention curve highlights the exceptional stability of Cr-Ce-MOF over 5000 cycles as illustrated in Figure 4.



Figure 6. Cyclic Voltammetry (CV)



Galvanostatic Charge-Discharge (GCD)











Discussion:

The results of this study underscore the remarkable potential of chromium-modified cerium metal-organic frameworks (Cr-Ce-MOF) as an advanced electrode material for supercapacitors. The superior electrochemical performance observed in this research is a direct consequence of the synergistic effects of chromium doping, the high surface area of the



MOF structure, and its robust porosity. These attributes collectively enhance the faradic reactions and ion transport, enabling efficient charge storage.

A key finding of this study is the high specific capacitance of the Cr-Ce-MOF electrode, which reached 646 F/g at 1 A/g. This value significantly exceeds the 423 F/g reported for undoped Ce-MOF in similar experimental conditions. The improvement can be attributed to the incorporation of chromium, which introduces additional redox-active sites within the framework, enhancing the faradic reaction processes. Comparatively, Gopalan et al. reported a capacitance of 523 F/g for hexagonal CeO₂. The Cr-Ce-MOF electrode, with its combination of high capacitance and excellent rate performance, demonstrates superior performance over these materials.

The galvanostatic charge-discharge (GCD) analysis further highlights the excellent rate capability of the Cr-Ce-MOF electrode. At a high current density of 10 A/g, the electrode retained 85% of its specific capacitance, a substantial improvement over the retention rates reported for other MOF-based materials. For instance, Padmanathan et al. observed 73% retention for carbon-supported CeO₂ nanorods under similar conditions. This enhancement is likely due to the enhanced conductivity and open pore structure of the Cr-Ce-MOF, which facilitates rapid ion transport and minimizes resistance.

Electrochemical impedance spectroscopy (EIS) results also confirm the superior conductivity of the Cr-Ce-MOF electrode. The Nyquist plot revealed a low charge transfer resistance of 0.72 Ω , which is lower than the values reported for comparable materials, such as Co-MOF-derived Co₃O₄ at 0.98 Ω . The steep slope of the low-frequency region further indicates efficient ion diffusion, a critical factor for maintaining high performance at varying charge and discharge rates.

Cycling stability is another critical parameter in evaluating the practicality of electrode materials. The Cr-Ce-MOF electrode demonstrated exceptional durability, retaining 92% of its initial capacitance after 5000 charge-discharge cycles at 5 A/g. This result surpasses the performance of CeO₂-graphene composites, which retained 85% capacitance after 3000 cycles as reported by Wang et al. The robust framework of Cr-Ce-MOF, combined with its resistance to structural degradation during cycling, ensures long-term reliability.

Overall, the incorporation of chromium into the cerium-based MOF framework not only enhances the redox activity but also improves the material's structural stability and conductivity. The high surface area, coupled with the material's ability to sustain performance under high current densities and prolonged cycling, makes Cr-Ce-MOF an outstanding candidate for next-generation supercapacitors. These findings align with the growing need for sustainable, high-performance energy storage systems and pave the way for further exploration of MOF-based materials in electrochemical applications.

Conclusion:

This study demonstrates the significant potential of chromium-modified cerium metalorganic frameworks (Cr-Ce-MOF) as electrode materials for supercapacitors. Chromium doping effectively enhanced the electrochemical performance by increasing the redox-active sites, conductivity, and porosity of the MOF framework. The synthesized Cr-Ce-MOF achieved a high specific capacitance of 646 F/g, exceptional rate capability with 85% retention at 10 A/g, and outstanding cycling stability of 92% retention after 5000 cycles at 5 A/g. These properties position Cr-Ce-MOF as a superior alternative to conventional Ce-MOFs and other MOF-derived materials. The study's findings underscore the importance of synergistic doping strategies to optimize the performance of MOF-based materials. The enhanced structural



stability and electrochemical properties of Cr-Ce-MOF pave the way for its application in highperformance energy storage devices. Future work could focus on scaling the synthesis process, exploring other metal combinations, and integrating these materials into hybrid energy storage systems for broader applications.

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