جامعة الملك عبدالله للعلوم والتقنية
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Exploring cathode materials for high performance water-based batteries

Toward Sustainable and High-Performance Zinc-Ion Energy Storage

Muruj Kassem
muruj.kassem@kaust.edu.sa

Mentor: Wandi Wahyudi
wandi.wahyudi@kaust.edu.sa

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Abstract

This study explores the electrochemical performance of various cathode materials for aqueous zinc-ion batteries (ZIBs), with a focus on vanadium-based compounds, particularly V_2O_3 and V_2O_5. Aqueous ZIBs offer a promising energy storage solution due to their safety, low cost, and environmental sustainability. The cathodes were fabricated and evaluated using cyclic voltammetry (CV), electrochemical impedance spectroscopy (EIS), and long-term galvanostatic cycling to assess redox activity, charge transfer resistance, and cycling stability. Among the tested materials, V_2O_3 exhibited superior performance, delivering higher capacity, enhanced stability, and lower impedance. V_2O_5 also demonstrated strong electrochemical activity under optimized formation and cycling conditions. The addition of $MnSO_4$ to the electrolyte significantly improved long-term cycling performance. In contrast, other cathode materials showed comparatively lower electrochemical performance, reinforcing the potential of vanadium oxides as leading candidates for next-generation high-performance water-based batteries.

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1 Introduction

This project was carried out by Muruj Kassem, under the supervision of Dr. Wandi Wahyudi and Prof. Husam N. Alshareef, with responsibilities covering material preparation, electrochemical testing, data analysis, and documentation. The motivation behind this work stems from the global demand for safer, more sustainable energy storage solutions. Aqueous zinc-ion batteries (ZIBs) have gained increasing attention due to their intrinsic safety, affordability, and the use of non-flammable, water-based electrolytes.

However, one of the major challenges limiting the commercialization of ZIBs is the availability of high-performance and stable cathode materials. This study hypothesizes that certain transition metal oxides and carbon-based electrodes can enhance Zn^{2+} storage capacity and improve the overall electrochemical stability in aqueous systems.

The objective of this project is to evaluate and compare the electrochemical performance of five different cathode materials: V_2O_5, V_2O_3, $NaNiFeMnO_2$ (NFM), MoO_3, and Kuraray activated carbon YP-50F. These materials were tested in aqueous electrolytes based on 3.0 M $ZnSO_4$, with and without the additive 0.5 M $MnSO_4$, using cyclic voltammetry (CV), electrochemical impedance spectroscopy (EIS), and galvanostatic cycling.

The results revealed that V_2O_3 delivered the highest capacity and cycling stability with low impedance, followed by V_2O_5 under optimized formation conditions. The presence of $MnSO_4$ further enhanced the cycling performance. By contrast, NFM, MoO_3, and carbon materials demonstrated lower electrochemical activity under the same conditions, reaffirming the promise of vanadium-based cathodes for next-generation water-based batteries.

2 System Overview

This study investigates the electrochemical performance of various cathode materials in aqueous zinc-ion batteries (ZIBs), a promising energy storage system known for safety, low cost, and environmental compatibility. The goal is to enable fair comparison of cathode materials under consistent conditions, allowing accurate assessment of their behavior in practical ZIB environments.

The cell architecture includes a zinc metal anode, a fiberglass separator, and a cathode coated on a stainless steel current collector. The cathode materials tested are V_2O_5, V_2O_3, $NaNiFeMnO_4$ (NFM), MoO_3, and Kuraray activated carbon (YP-50F), selected based on redox activity, structural stability, and electrolyte compatibility.

The electrolyte used is 3.0 M $ZnSO_4$, with additional tests using 0.5 M $MnSO_4$ as an additive to evaluate its effect on cycling and ion diffusion.

Electrochemical testing included:

- Cyclic Voltammetry (CV) to analyze redox activity
- Electrochemical Impedance Spectroscopy (EIS) to evaluate charge transfer resistance
- Galvanostatic cycling to assess capacity and cycling stability

A schematic representation of the Zn-ion battery setup is provided below.

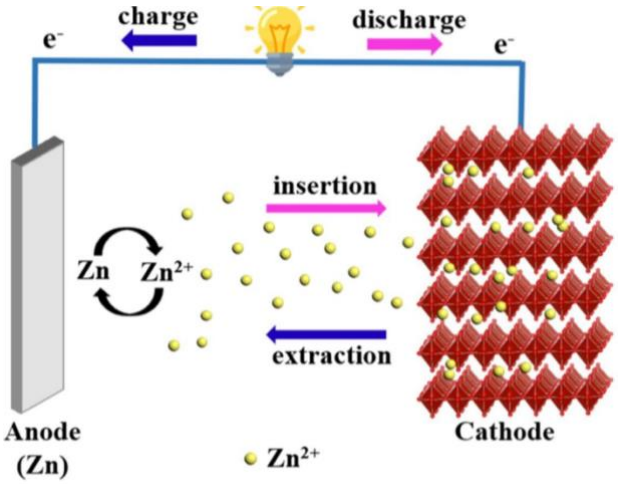


Figure 1. Schematic diagram of an aqueous Zn-ion battery showing electron flow, Zn^{2+} migration, and electrode roles during charge–discharge cycles.

3 Methodology

3.1 Cathode Preparation

Cathode slurries were prepared using a mixture of active material (0.8 g), Super P conductive carbon (0.1 g), and PVDF binder (2.5 g) in an 8:1:1 weight ratio. Twelve drops of N-methyl-2-pyrrolidone (NMP) were added as the solvent to form a uniform slurry. The slurry was mixed using a conditioning mixer for 8 minutes. The slurry was then coated onto a stainless steel sheet (serving as the current collector) using a 300 µm film applicator. The coated electrodes were dried in a vacuum oven at 80 °C for 50 minutes. After drying, the electrodes were subjected to plasma sputtering treatment to improve surface wettability and promote better contact between the cathode and the aqueous electrolyte. The final electrodes were punched into 14 mm diameter discs.



Figure 2. Cathode fabrication steps, including mixing of materials, slurry preparation, coating, drying, and electrode punching.

3.2 Cell Assembly

Coin-type (CR2032) cells were assembled using the prepared 14 mm cathodes, 14 mm zinc metal anodes, and 18 mm fiberglass separators. Two types of aqueous electrolytes were used:

- 3.0 m $ZnSO_4$
- 3.0 m $ZnSO_4$ + 0.5 m $MnSO_4$

A fixed volume of 120 µL of electrolyte was added to each cell. Cells were assembled for five cathode materials: V_2O_5, V_2O_3, $NaNiFeMnO_2$ (NFM), MoO_3, and YP-50F activated carbon.

3.3 Electrochemical Testing

Electrochemical performance was evaluated using:

- Cyclic Voltammetry (CV) to assess redox activity.
- Electrochemical Impedance Spectroscopy (EIS) to evaluate charge transfer resistance.
- Galvanostatic charge–discharge cycling to determine specific capacity and cycling stability.

CV and EIS measurements were conducted using a laboratory-grade potentiostat/galvanostat (manufacturer unspecified), while galvanostatic cycling was performed using a Neware battery testing system. All electrochemical tests were conducted at room temperature. A formation step at low current density was applied before long-term cycling to stabilize the electrode–electrolyte interface. For most materials, a fixed current density of 2 A/g was used for cycling after formation. However, for V_2O_5, different formation and cycling current combinations were explored to optimize electrochemical performance:

- 0.01 A/g formation + 2 A/g cycling
- 0.05 A/g formation + 2 A/g cycling
- 0.05 A/g formation + 0.2 A/g cycling

These varying conditions were used to investigate the influence of pre-conditioning on capacity retention and stability. All electrochemical results were used to compare the performance of different cathode materials under consistent aqueous conditions.

4 Results and Discussion

This section presents the electrochemical evaluation of five different cathode materials (V_2O_5, V_2O_3, MoO_3, NFM, and Kuraray carbon) in aqueous zinc-ion coin cells. The analysis includes galvanostatic charge/discharge cycling, cyclic voltammetry (CV), and electrochemical impedance spectroscopy (EIS), focusing on capacity retention, redox behavior, and internal resistance.

4.1 Electrochemical Cycling Performance

Long-term cycling tests were conducted over 1000 cycles. V_2O_3 showed the highest discharge capacity, maintaining values above 100 mAh/g with excellent cycling stability. V_2O_5 demonstrated a continuous capacity rise during cycling, stabilizing around 120 mAh/g. The gradual improvement indicates potential activation processes within the electrode.

MoO_3, NFM, and Kuraray carbon displayed low and relatively flat capacities, typically below 50 mAh/g. These results suggest weak Zn^{2+} intercalation activity or limited conductivity for these materials under the tested conditions.

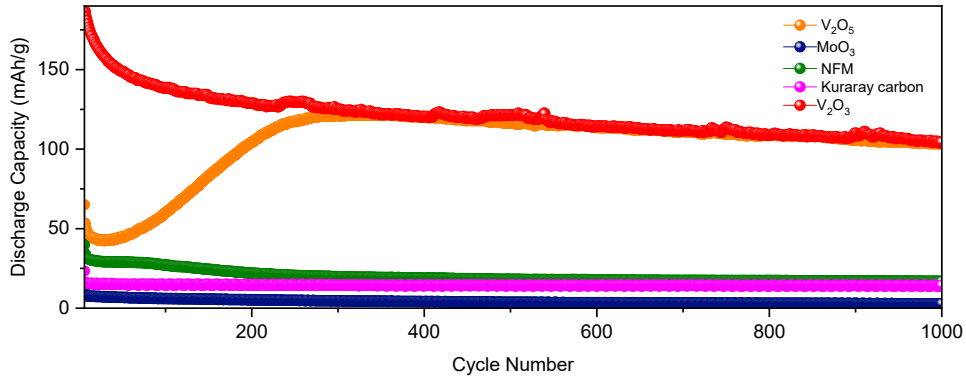


Figure 3. Long-term cycling performance of various cathode materials in aqueous Zn-ion batteries.

These outcomes align with prior findings where vanadium oxides tend to outperform other metal oxides in aqueous ZIBs due to their layered structure and favorable redox chemistry.

4.2 Impact of Formation Conditions and Electrolyte Composition (V_2O_5 Study)

To optimize V_2O_5 performance, a series of tests were conducted by varying the formation current, cycling current, and electrolyte formulation.

Cells formed at a lower current density (0.01 A/g) displayed significantly higher capacity and improved stability compared to those formed at 0.05 A/g. Furthermore, using a mixed electrolyte (3.0 m $ZnSO_4$ + 0.5 m $MnSO_4$) enhanced capacity retention and cycling performance relative to the $ZnSO_4$-only electrolyte.

These results suggest that both gentle pre-conditioning and electrolyte modification are critical for activating the V_2O_5 structure and enhancing Zn^{2+} transport and interfacial stability.

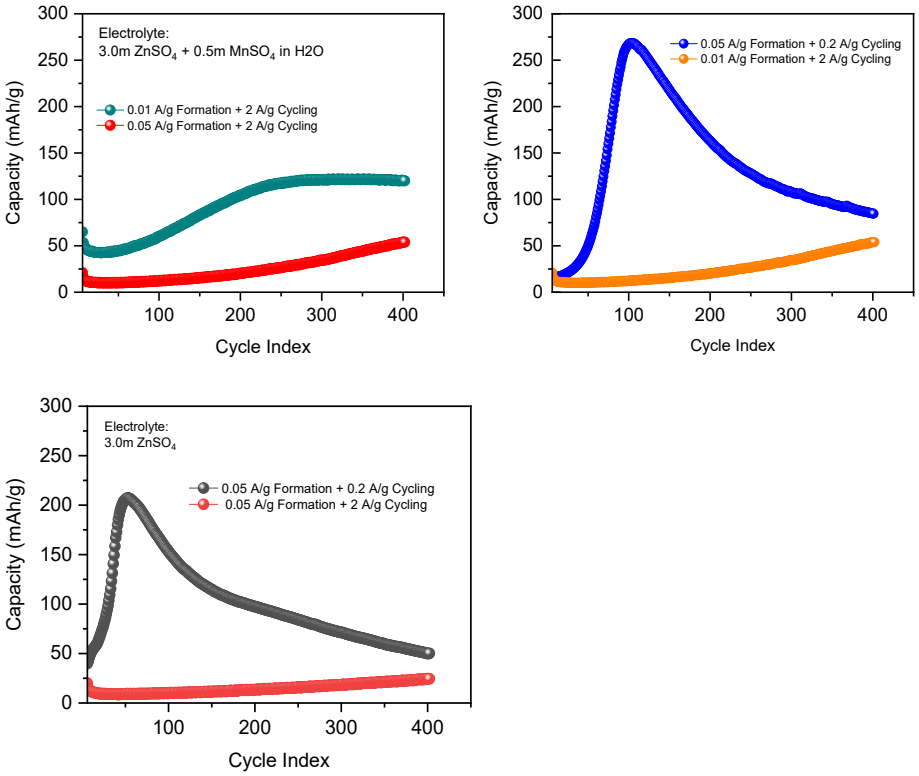


Figure 4. Effect of formation current and Mn^{2+} electrolyte additive on V_2O_5 cycling behavior.

4.3 Cyclic Voltammetry (CV) Analysis

CV scans were recorded to analyze redox activity and reversibility of each cathode.

V_2O_3 showed pronounced redox peaks with strong symmetry, indicating highly reversible redox processes. V_2O_5 and MoO_3 exhibited moderate redox features, while NFM showed broad, poorly defined peaks, reflecting slow kinetics or incomplete redox conversion.

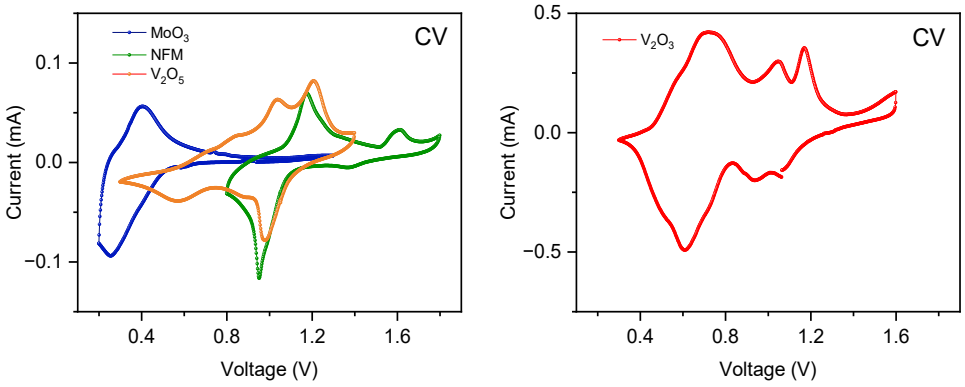


Figure 5. Cyclic voltammetry results for different cathodes tested in aqueous Zn-ion cells.

The peak areas were generally consistent with the observed discharge capacities. Reversibility and peak sharpness correlated well with the cycling performance.

4.4 Electrochemical Impedance Spectroscopy (EIS)

EIS was used to measure charge transfer resistance before and after CV cycling.

Prior to CV, V_2O_3 had the lowest impedance, suggesting excellent electrode kinetics. NFM showed the highest resistance, which may explain its poor capacity. V_2O_5 and MoO_3 exhibited moderate impedance values.

Following CV, V_2O_5 displayed a noticeable reduction in impedance, attributed to improved electrode wetting and electrochemical activation. Conversely, MoO_3 showed a sharp increase in resistance, suggesting material degradation. NFM remained high, and V_2O_3 retained low impedance, consistent with its superior performance.

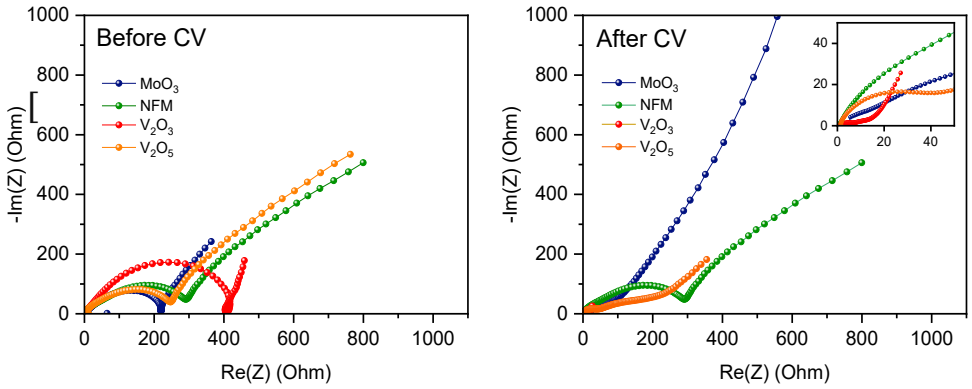


Figure 6. Electrochemical impedance diagrams for the cathodes before and after CV testing.

4.5 Performance Summary and Recommendations

- Best performer: V_2O_3 exhibited the most stable and highest capacity performance across all evaluations.
- Electrolyte engineering: Addition of $MnSO_4$ significantly enhanced capacity in V_2O_5-based cells.
- Optimization insight: A low formation current (0.01 A/g) proved essential for stable cycling.
- Underperforming cathodes: NFM and MoO_3 showed limited practical performance and may require further material modification or compositing.
- Method success: Plasma sputtering effectively enhanced cathode–electrolyte interface, as confirmed by EIS improvement and capacity increase.

5 Reflections on Challenges

Throughout the experimental work, several challenges were encountered and gradually resolved. One of the early technical issues was controlling the slurry consistency and determining the appropriate amount of NMP solvent. It took several trials to find a balanced mixture that ensured smooth coating and stable electrode performance.

Another key challenge was optimizing the ratio between the active material, conductive carbon, and binder. After testing different compositions, the ratio was optimized and fixed at 8:1:1, which provided the best results in terms of electrode structure and conductivity.

Optimizing the current used for both the formation and cycling steps was also a major focus. Multiple combinations of formation and cycling currents were tested, especially for V_2O_5. It was found that using a low formation current of 0.01 A/g followed by a 2 A/g cycling current resulted in significantly better capacity retention and stability over extended cycles.

In addition, poor wettability between the electrode surface and the aqueous electrolyte was observed in some samples, which negatively impacted the initial electrochemical performance. This issue was effectively addressed by applying a plasma sputtering treatment to the cathode surface, which improved contact with the electrolyte and enhanced overall cell performance.

6 Conclusion and Future Work

This study investigated the electrochemical performance of five different cathode materials for aqueous zinc-ion batteries. Among them, vanadium-based oxides, particularly V_2O_3 and V_2O_5, showed superior performance in terms of capacity, cycling stability, and lower resistance. V_2O_3 exhibited the best overall performance, while V_2O_5 showed strong redox activity and improved significantly under optimized cycling conditions. In contrast, MoO_3, NFM, and activated carbon displayed weaker electrochemical behavior.

Key performance improvements were achieved through the optimization of formation and cycling currents, where a formation current of 0.01 A/g followed by 2 A/g cycling led to enhanced capacity and stability for V_2O_5. Additionally, plasma sputtering treatment effectively improved electrolyte wettability on the electrode surface, contributing to better interfacial contact and more stable cycling.

Despite these promising results, one major limitation of this study is that all cathode materials were used in their commercial form without structural engineering. However, it is well established that the crystal structure, particle size, and morphology of vanadium oxides play a crucial role in determining their electrochemical behavior.

Future work will focus on the synthesis of nanostructured vanadium oxides with optimized properties, such as porous frameworks, layered morphologies, or nanoscale architectures. These tailored materials are expected to provide faster ion transport, greater surface area, and improved structural stability—further enhancing the performance of aqueous zinc-ion batteries and supporting their development for practical energy storage applications.

7 References

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