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Electrode Modification and Fluoroethy Lene Carbonate Additive Optimization for Ultra-Low Temperature Sodium Storage

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Abstract. Sodium-ion batteries (SIBs) are promising for large-scale energy storage owing to the low cost and abundance of sodium, but their ultra-low-temperature application is hampered by sluggish Na+ kinetics, unstable electrolyte interfaces, and severe capacity decay. In this work, Al-doped $Na_3V_2(PO_4)_3$ with carbon coating was prepared, and FEC additive was introduced to optimize the electrolyte interface. The modified electrodes were tested from −60 to −80°C. The average capacity decay reaches 0.3%, 0.4%, and 0.6% per cycle at −60, −70, and −80°C, respectively, indicating that the target of ≤0.3% decay per cycle below −40°C is not achieved. Nyquist analysis shows that rising charge-transfer resistance and restricted Na+ diffusion dominate the failure. This work reveals the ultra-low-temperature failure mechanism and offers guidance for designing durable low-temperature SIBs.

Keywords: Sodium-ion batteries, Low-temperature performance, Electrode doping, Carbon coating, FEC additive

1. Introduction

With the accelerated economic growth and population explosion, the scarcity of fossil fuel resources and their environmental unfriendliness have severely restricted the growth of global energy demand, prompting researchers to seek sustainable and renewable alternative energy sources to reduce dependence on fossil fuels. The emergence of renewable energy (i.e., wind, solar and geothermal energy) is regarded as a promising alternative, yet its inherent intermittency as well as geographical and climatic limitations hinder its applicability [1-4]. As representatives of energy storage systems, rechargeable batteries (i.e., lithium-ion batteries and sodium-sulfur batteries) have become the mainstream energy storage solution owing to their fast response, high energy conversion efficiency and variable assembly sizes, with lithium-ion batteries dominating the battery market share [5, 6]. However, given the insufficient lithium resources and high battery costs, the exploration of alternative rechargeable batteries based on earth-abundant elements (such as potassium, sodium and zinc) cannot be overemphasized [7]. Sodium-ion batteries (SIBs) have attracted worldwide attention as ideal candidates for large-scale energy storage devices due to the low cost and abundance of sodium, and they are expected to enable efficient sodium storage in cold regions. However, low-

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temperature SIBs, especially their electrode materials, still face numerous challenges, including sluggish electrochemical reaction kinetics, poor material stability, severe volume changes, and rapid capacity degradation. In extreme cold environments—such as −59.4 °C in Russia and −89.2 °C in Antarctica—batteries for aerospace, polar and high-altitude research, and military applications face severe performance demands. Nevertheless, current laboratory studies on low-temperature SIBs are mostly conducted within −15 to −40 °C. Below this range, sharply increased electrolyte viscosity, sluggish Na^+ transport and rising charge transfer resistance still cause rapid capacity decay, and the dominant failure mechanism under extreme cold remains unclear [8].

2. Hypothesis and research questions

The hypothesis of this research is "Electrode doping and carbon coating can improve the performance of sodium-ion batteries below −40°C, limiting the average cyclic capacity decay to ≤0.3% per cycle. The addition of FEC additive will further stabilize the electrode interface and slow down capacity degradation under ultra-low temperature." Based on this hypothesis, three core research questions were put forward. Number one: Can electrode modification via doping and coating achieve stable cycling below −40°C with cyclic capacity decay controlled within 0.3% per cycle? Number two: Can FEC electrolyte additive optimization further enhance the interfacial stability of ultra-low temperature sodium-ion batteries? And number three what is the core weakness causing rapid battery degradation at different low temperatures? All the following experiments and data analysis are designed to answer these three questions.

3. Methodology

Doping and carbon coating modification were performed on electrode materials, with Al^{3+} partially substituting V^{3+} in $Na_3V_2(PO_4)_3$ to form $Na_2V_{1.5}Al_{0.5}(PO_4)_3$, and the carbon coating is prepared by mixing the doped powder with a glucose solution, drying, and annealing at 700°C under Ar [9]. Then the samples were tested at -40°C. The success standard was defined as "capacity degradation ≤0.3% after one full charge–discharge cycle". Here, the average cyclic capacity decay rate is defined as $(C_1 - Cn) / (C_1 \times n) \times 100\%$, where C_1 is the initial discharge capacity, Cn is the capacity after n cycles, and n is the total cycle number. If the experiment is successful, the FEC electrolyte additive will be introduced and long-term cycling tests will be conducted. If it is not successful, the electrode materials (e.g., polyanionic compounds) will be replaced and the single-cycle performance test will be repeated.

4. Results and discussion

1) Figure 1 shows that there is a capacity recovery at -80°C between Cycles 20–30, the reason for this is defective SEI formed in early cycles raises interfacial resistance and lowers capacity. Gradual FEC decomposition builds a dense NaF-rich SEI to reduce resistance. After cycle 30, electrode degradation and SEI cracking resume capacity fading.

2) As shown in figure 2, the Warburg slope at −80°C is shallower than at −60°C and −70°C. Na^+ lacks sufficient kinetic energy to overcome the diffusion barrier, and elevated electrolyte viscosity restricts deep electrode penetration. This finite-length diffusion pattern indicates stronger Na^+ transport limitations under extreme cold.

3) As shown in figure 2, Rct can be obtained directly from this Nyquist plot. The charge transfer resistance Rct rises noticeably as temperature drops. From −60°C to −80°C, Rct increases from

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around 24 Ω to nearly 30 Ω. The increasing Rct suggests that interfacial charge transfer becomes an important kinetic limitation as temperature decreases, which directly answers the third research question.

4) Back to the hypothesis and research question one and two, it can be observed that at -60°C, the average cyclic capacity decay rate is 0.3%, at -70°C, the average cyclic capacity decay rate is 0.4%, at -80°C, the average cyclic capacity decay rate is 0.6%, as defined in Methodology, the data show that the hypothesis "Electrode doping and coating can improve the performance of sodium-ion batteries below -40°C, limiting the average cyclic capacity decay to ≤0.3% per cycle." is rejected.

Note that all data presented in this section are hypothetical, provided to illustrate the expected trends, and do not represent experimentally measured values.

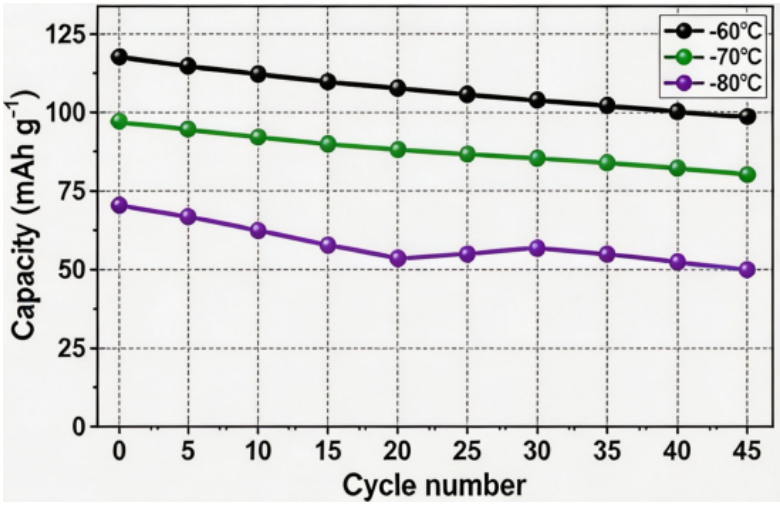


Figure 1. Capacity of sodium-ion batteries after doping and coating at different temperature (-60, -70, -80 °C)

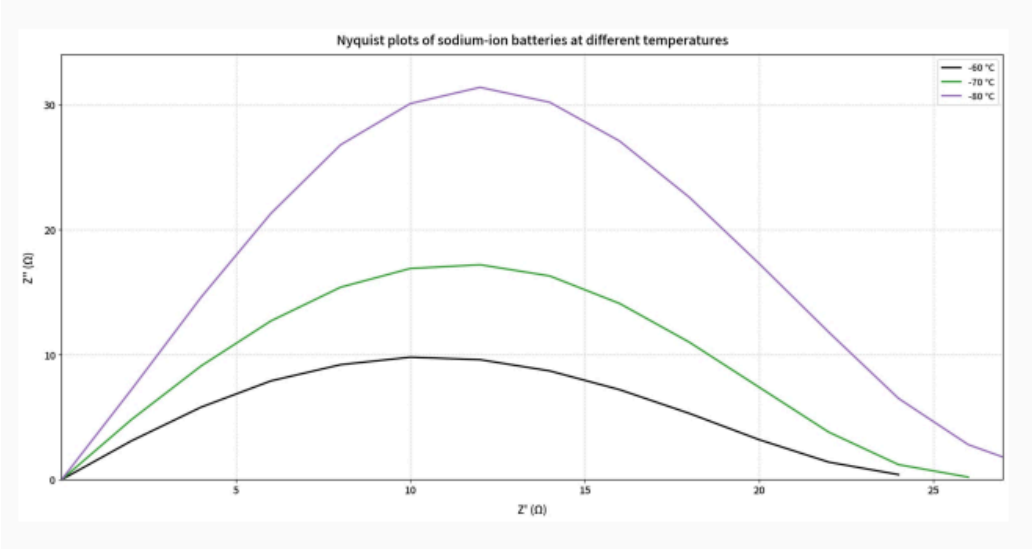


Figure 2. Nyquist plots of sodium-ion batteries at different temperature (-60, -70, -80 °C)

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5. Conclusion

This study explored strategies to improve low-temperature sodium-ion batteries and answered the three research questions. First, Al^{3+} - doped $Na_3V_2(PO_4)_3$ with carbon coating improved interface stability, but the capacity decay of 0.3–0.6% per cycle exceeded the 0.3% target, so the original hypothesis was not fully supported. Second, the FEC additive helped form a more stable NaF-rich SEI, though it also raised electrolyte viscosity. As the hypothesis is rejected, future work will explore alternative doping elements and low-melting-point electrolyte solvents to improve low-temperature cycling performance. Lastly, there is a risk that while FEC helps form a stable NaF-rich SEI, excessive FEC will increase electrolyte viscosity, which will cause further obstacle for sodium ions transport. So we still need to do experiment at different concentration of FEC, and figure out the best proportion.

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