Architectured Synthesis of a NMC core/shell cathode: Tailoring microstructure with tungsten oxide barrier for improved stability of Lithium- ion batteries

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Lithium-ion batteries play a crucial role in modern energy storage applications, driven by the demand for high-performance and widely applicable energy storage solutions. High Ni-content layered oxide cathodes, such as $LiNi_xMn_yCo_zO_2$ (NMC) applied in Lithium-ion batteries (LIBs), are at the forefront of next-generation cathode materials due to their high energy density and cost-effectiveness. However, their commercialization is hindered by several challenges, including irreversible capacity loss during the initial cycle, voltage hysteresis, poor rate capability, and gradual voltage decay. These issues primarily stem from the high Ni-content, which promotes Ni^{2+} migration into vacant lithium sites via tetrahedral interstices after Li^+ extraction. This migration is accompanied by oxygen lattice loss, leading to structural instability and performance degradation [1]. To address these challenges, we propose a core/shell NMC architecture, where a high-Ni NMC90 core ensures high capacity, while a Mn-rich NMC622 shell enhances structural and thermal stability [2]. Furthermore, introduction of a tungsten oxide (WO_3) interlayer between the core and shell may offer additional benefits to mitigate interfacial degradation and improve long-term cycling stability. This WO_3 layer acting as a diffusion barrier may prevent undesirable elemental migration and preserve the structural and electrochemical integrity of the cathode material. The synthesis of this multi-layered core/shell NMC particles has been performed using a co-precipitation method, employing oxalic acid as a cost-effective and environmentally friendly precipitant. Additionally, lithium incorporation has been carried out directly during the co-precipitation step to ensure a homogeneous precursor composition. Structural and electrochemical characterizations have been conducted using SEM, EDX, XRD and GCD analysis. The XRD results in Figure 1(a) confirm that both NMC90 and NMC622 achieve the desired $R\bar{3}m$ crystal structure on calcination at 850°C. Furthermore, GCD measurements, shown in Figure 1(b), reveal that the WO_3-modified core/shell cathode exhibits a degradation rate that is reduced by a factor of over two compared to the uncoated counterpart during cycling. This combined strategy of integrating a core/shell architecture with a WO_3 diffusion barrier, demonstrates a promising approach for enhancing the performance, stability, and lifespan of high-Ni NMC cathodes in next-generation LIBs.

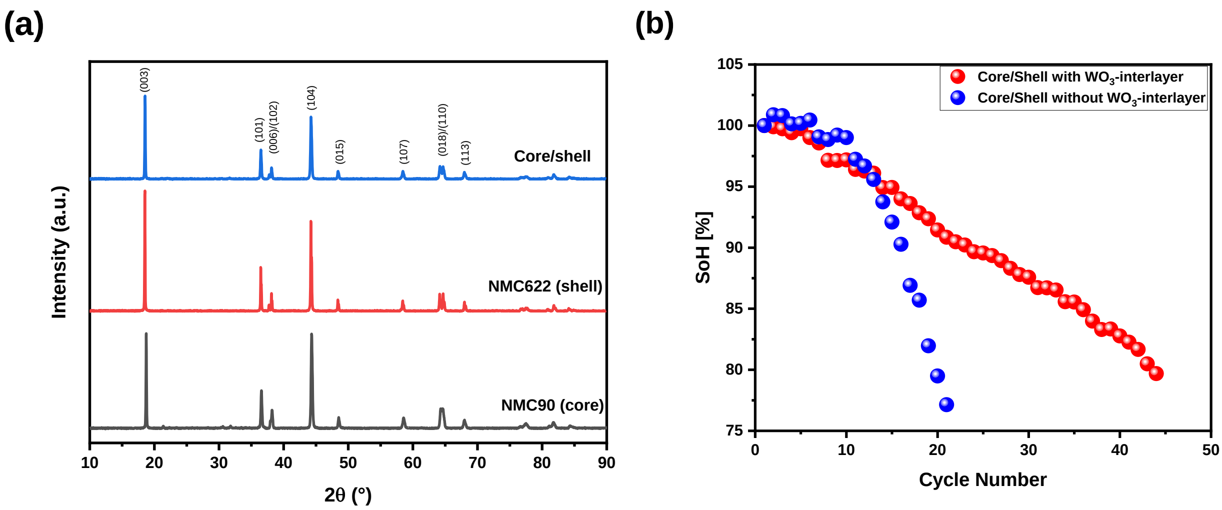

Figure 1: XRD of NMC90 (core), NMC622 (shell) and core/shell structured NMC cathode material and (b) cycling performance of core/shell structured NMC with and without WO_3 Interlayer

REFERENCES

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