Development of NMC cathode powders with optimized core/shell architecture

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High-nickel $LiNi_xMn_yCo_zO_2$ (NMC) cathode materials are increasingly becoming popular for their potential to increase storage capacity, reduce cost and environmental impact. A key objective is to push the specific capacity of these materials as close as possible to the theoretical limit of $LiNiO_2$ (LNO), which is 275 mAh/g, while simultaneously ensuring long-term cycling stability beyond 1000 cycles [1]. However, increase of the Ni content in cathodes presents several challenges, including rapid capacity fading and poor thermal stability. These issues originate from the high reactivity of Ni with surface oxygen during charge-discharge cycles, as well as significant Li/Ni cationic mixing resulting in degradation of electrochemical performance [2]. The priority aim, being the enhancement of structural and thermal stability and elimination of these limitations, has been addressed through the synthesis of core/shell-structured cathode powder, featuring a Ni-rich NMC90 core for high specific capacity and a Mn-rich NMC622 shell. Hereby, a cost-effective and process controlling oxalate-assisted co-precipitation method has been employed to achieve the desired morphology. Various synthesis parameters such as calcination time, temperature and lithium content have been optimized to tailor the core/shell architecture. Structural, compositional, and electrochemical properties have been analysed using SEM, EDX, XRD, and GCD techniques. XRD results indicate that the $R\bar{3}m$ phase is obtained for both NMC90 and NMC622 compositions on calcination at 850 °C, as shown in Figure 1(a). The in-situ Li-infiltration approach leads to a relatively high I(003)/I(104) ratio, indicative of improved structural ordering. The core/shell structured NMC cathode also results in a higher specific capacity compared to the core and shell individually, as the interplay between both components creates a synergistic improvement. While the higher lithium content in the core further enhances specific capacity, the shell plays a crucial role in stabilizing the material during cycling. This combination optimizes the balance between capacity and long-term performance.

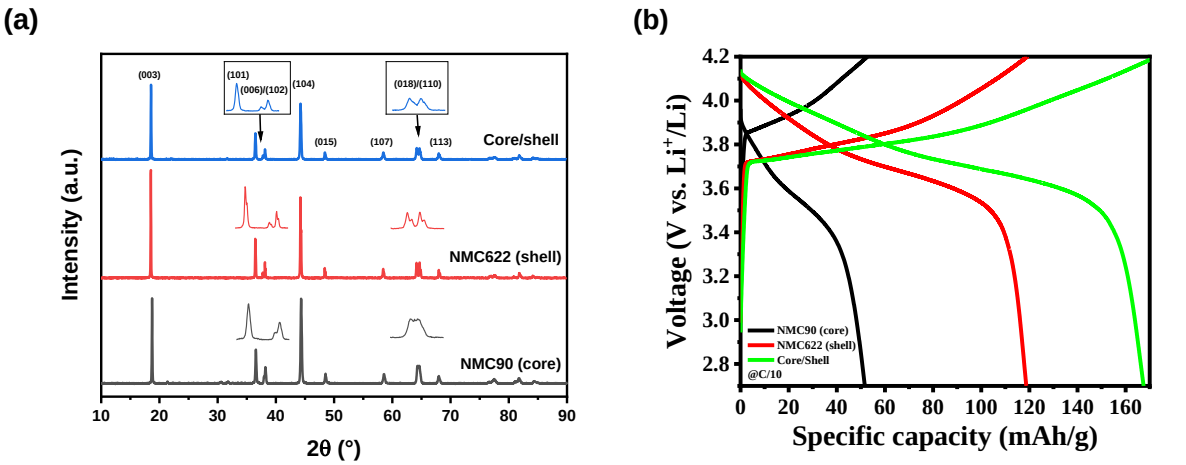

Figure 1: (a) XRD patterns of NMC90 (core), NMC622 (shell) and the core/shell cathode material and (b) charge-discharge voltage profile of these materials

References

[1] D. Weber et al., *Eur. J. Inorg. Chem.*, 2020, 33, 3117-3130
[2] H. R. Oliveira Filho et al., J. Energy Storage, 2024, 82, 110536