

Development of a wet-chemical Al₂O₃ coating synthesis on LiNi_{0.33}Co_{0.33}Mn_{0.33}O₂ (NCM111) electrode material for cycle life extension of Li-ion batteries

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Impact on the environment from lithium-ion battery (LIB) disposal can be lowered not only by developing greener recycling methods, but, perhaps even more effectively, by eliminating the need to dispose of batteries so often in the first place. Therefore, extension of LIB lifetime is an important research direction for the future of mobile electronics.

LIB degradation can be thought of on multiple levels. Overall, the degradation mechanisms causing capacity fade over time can be divided into loss of lithium inventory (LLI) and loss of electrochemically active material (LAM). Going into more detail, the most significant effect on overall battery degradation stems from cathode degradation-related processes. Main degradation pathways of the state-of-art LiNi_xCo_yMn_{1-x-y}O₂ (NCM) cathode material include surface phase transitions with associated oxygen release, particle cracking due to volume changes during charge-discharge cycling, transition metal (TM) dissolution, and cathodic solid-electrolyte interphase (CEI) growth.¹ These degradation pathways contribute to either LAM from the cathode, LLI, or growth of internal resistance. Furthermore, cathodic degradation can lead to other types (electrolyte or anode-related) of degradation. For example, the oxygen evolved during phase transitions can detrimentally react with the electrolyte and produce CEI-forming species. Preventing cathode degradation would alleviate a large share of degradation within the battery.

Among doping and structural control, coating the cathode material is one of the most effective ways to mitigate cathode degradation. Coatings can be formed by various methods, like atomic layer deposition, chemical vapor deposition, and others, among which the wet-chemical method shows the most promise for relatively fast, cost-effective, and easy up-scaling.

In this work, an inert Al₂O₃ coating is synthesized on LiNi_{0.33}Co_{0.33}Mn_{0.33}O₂ (NCM111) active material by an easily up-scalable wet-chemical method based on a non-toxic coating precursor (aluminium isopropoxide or AIP) and solvent (ethanol). The coated material is characterized by various structural analysis methods (SEM, TEM, XRD, XPS), and LIB half cells containing the coated material are assembled for rate capability and cyclability testing. A significant improvement in capacity retention is observed for the coated material. Additionally, an insight into the effects of the coating procedure itself (without adding the AIP precursor) is given for a deeper understanding on the origin of improved cycling stability. The washing and sintering steps of the wet-chemical coating procedure were investigated separately and together. It was revealed that washing NCM111 in ethanol is detrimental for capacity retention, whereas sintering the active material in air improves the initial capacity and has slight deleterious effect on cycling stability. However, washing and sintering together (reference material) has a positive effect on both cycling stability and capacity retention. Nevertheless, the washing and sintering procedure alone cannot substitute coating with Al₂O₃, as the cycling stability is even further improved for the coated material, showing a capacity retention of 88% (vs. 79% for the reference material) after 500 charge-discharge cycles at 1C in the voltage range 2.7-4.3 V vs. Li/Li⁺.

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References

1 L. Britala, M. Marinaro and G. Kucinskis, *Journal of Energy Storage*, 2023, **73**, 108875.