



# Spray-drying Al onto hydroxide precursors to prepare $\text{LiNi}_{0.855}\text{Co}_{0.095}\text{Al}_{0.05}\text{O}_2$ as a highly stable cathode for lithium-ion batteries



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## ABSTRACT

The very high specific discharge capacities of Ni > 90% cathodes are often undone by their extremely poor cycle life and thermal stability. Herein, Ni-poor, and Al-rich particle with double concentration gradients is fabricated by synthesizing particles of the Ni-rich hydroxide  $\text{Ni}_{0.9}\text{Co}_{0.1}(\text{OH})_2$  in a highly efficient Taylor flow (TF) reactor and then depositing Al onto their surfaces using three methods: spray-drying and conventional wet and dry chemical coating. The uniformity of the surface distribution of Al on the Ni-rich transition metal oxide cathode materials affected the overall structural and electrochemical stability. Among our three systems, the cathode material prepared with the spray-dried Al exhibited the best performance, with an initial discharge capacity of  $196.9 \text{ mA h g}^{-1}$  and capacity retention of 93% after 100 cycles at a rate of 1 C. It also demonstrated superior electrochemical performance at higher C-rates. For example, at 10 C, it delivered an initial discharge capacity of  $134.4 \text{ mA h g}^{-1}$ , compared with  $56.8 \text{ mA h g}^{-1}$  for the Al-free  $\text{LiNi}_{0.9}\text{Co}_{0.1}\text{O}_2$ . We attribute this enhanced electrochemical performance to the presence of Al and its uniform distribution (through spray-drying processing) on the outer layer of the active material, with the Ni and Co elements remaining mainly within the core.

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## 1. Introduction

Lithium-ion batteries (LIBs) are in extremely high demand for energy storage, with applications ranging from portable electrical and electronic gadgets to transportation [1]. Electric vehicles (EVs) account for a substantial share of the LIBs, with this market on track to explode from 4 million EVs on the road in 2018 to a lower estimate of 50 million EVs and a higher estimate of 225 million EVs by 2030 [2,3]. As a result, significant amounts of research, development, and engineering have been directed toward the creation of lower-cost, higher-energy-density LIBs [4,5].

Layered  $\text{Li}[\text{Ni}_x\text{Co}_y\text{Mn}_{1-x-y}]\text{O}_2$  (NCM) and  $\text{Li}[\text{Ni}_x\text{Co}_y\text{Al}_{1-x-y}]\text{O}_2$  (NCA) cathodes have already replaced  $\text{LiCoO}_2$  (LCO) cathodes in the LIBs for the EV market [6]. With the introduction of the Tesla New Energy Vehicle, which uses NCA as the cathode material, a significant

amount of research is being conducted into NCA cathodes [7]. Increasing the Ni content in NCM and NCA cathodes has resulted in considerable improvements in the LIB energy densities. Because the weaker Ni–O bonds disrupt the crystal structure and increase the anisotropic volume shift during Li extraction/insertion, this increase in Ni content is generally accompanied by sacrifices in the cycling and thermal stabilities. The specific capacity of an NCA cathode grows roughly proportionately to the Ni content for values of  $x$  of greater than 0.9, but the capacity gain from Ni enrichment is negated by the rapid capacity fading that occurs concurrently. As a result, most researchers have focused recently on tuning the balance between high capacity and quick fading.

To overcome the degradation of NCA electrodes, many investigations have focused on structural evolution, especially regarding changes in the surface structure during the initial and subsequent cycles. Cation mixing is one of the main phenomena that occur during synthesis, as well as during  $\text{Li}^+$  ion intercalation, due to the similar ionic radii of  $\text{Li}^+$  and  $\text{Ni}^{2+}$  ions. Increases in the temperature, the state of charge, and the Ni concentration in an NCA electrode can all promote cation mixing [8,9]. In addition to lowering

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