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A hybrid cathode, $\text{Li}[\text{Ni}_{0.886}\text{Co}_{0.049}\text{Mn}_{0.050}\text{Al}_{0.015}]\text{O}_2$, consisting of a core of $\text{Li}[\text{Ni}_{0.934}\text{Co}_{0.043}\text{Al}_{0.015}]\text{O}_2$ encapsulated by $\text{Li}[\text{Ni}_{0.844}\text{Co}_{0.061}\text{Mn}_{0.080}\text{Al}_{0.015}]\text{O}_2$ is prepared. This core/shell-type structure combining a Ni-enriched Li[NixCoyAl1-x-y]O2 (NCA) cathode with an Al-doped Li[Ni_xCo_yMn_{1-x-y}]O₂ (NCM) cathode provides an exceptionally high discharge capacity of 225 mAh g⁻¹ at 4.3 V and 236 mAh g⁻¹ at 4.5 V. The hybrid cathode also exhibits microstructural attributes that are beneficial to long-term cycling stability, namely, spatially correlated peripheral primary particles that are crystallographically textured to expedite Li intercalation and nano-sized core primary particles retard the propagation of interparticle microcracks. In addition, ordered intermixing of Li and transition metal ions is observed in the cycled hybrid cathode. This cation ordering stabilizes the host structure during cycling and facilitates Li intercalation. These structural features allow the hybrid cathode to retain 91% of its initial capacity after 1000 cycles, which easily surpasses the performance of currently available cathodes.