Stress-dissipation strategy stabilized solid oxide cathodes for high-performance allsolid-state lithium batteries

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The electrochemical-mechanical instability of single-crystalline Ni-rich oxide cathode associated with stress variation during lithium (de)lithiation poses a substantial barrier to the operation of high-energy density and fast charging in the all-solid-state lithium batteries (ASSLBs). Here we report an approach that suppresses the electrochemicalmechanical degradation using the integration of an innovative stress dissipation into Nirich oxides, which involves a F-doped bulk structure and island-like sticky SiO₂ gel on the surface of Ni-rich oxides through a simple washing process. F ions realize a stable interlayer structure manifested in its strong interactions with transition metals and oxygen, effectively constraining detrimental structure deformation to retardate stress redistribution during (de)lithiation. The island-like sticky gel clusters allow local stress gradient formation on the surface to drive the particle rotation, responsible for the main stress dissipation during long-term cycling. The oxide cathode design enables the sulfidebased ASSLBs to achieve a high specific capacity of 192 mAh g^{-1} on charging at 0.1 C while still maintaining 45 mAh g^{-1} at 10 C. Notably, the assembled sulfide-based ASSLB demonstrates superior cycling stability over 1000 cycles with 98 % capacity retention at 2 C. This generalizable approach offers insights into the stress dissipation path on the electrochemical-mechanical stability of solid oxide cathodes, opening up a new avenue for ASSLBs to achieve high energy density, fast charging capability, and long durability.



Figure 1. Electrochemical performance of ASSLBs at 30°C. a, Electrochemical rate capabilities. b. Galvanostatic discharge profiles of 83125 and SF-83 cathodes at various current rates. c. Long-term cycling performance of SF-83 at 2 C rate; d, Comparison of rate capability between the all-solid-state batteries assembled with SF-83 and previously reported Ni-rich oxide cathodes