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Motivated by the recent growth of electric vehicles (EVs), there is a significant worldwide effort focused on



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the development of safer and higher energy density solid-state batteries (SSBs)1,2. In SSBs, safety is enhanced by replacing the flammable liquid electrolyte (LE) with a non-flammable ceramic solid electrolyte (SE)2,3. Furthermore, SSBs have the potential to enable high-energy-density electrode materials including Li-metal anodes and high-voltage cathodes4,5,6,7,8,9,10,11.

To improve the electrochemical performance of NMC CAMs, surface coatings and doping are two commonly employed methodologies. These approaches have been shown to reduce the undesirable interactions at the CAM/electrolyte interface, as well as helping to mitigate irreversible phase transformations and maintain the structural integrity of the CAM14,21,26,27. Surface coatings on NMC can lead to a more stable CAM/electrolyte interface and suppress unwanted interactions, resulting in improved Coulombic efficiency. Additionally, by doping a different element into the NMC crystal structure, cation intermixing can be suppressed and the transitional metal (TM) layer spacing can be preserved21,26.

The procedure for depositing amorphous Nb2O5 coatings onto SC-NMC particles using ALD is depicted schematically in Figs. 1A and S1. ALD was performed on SC-NMC particles (sized 2-5 um) without any additional pretreatment. To ensure conformal coverage of the entire particle surface without the presence of discontinuities at particle-particle contact points, a rotary-bed ALD reactor was used (Figs. 1A and S1)45,46. In this process, the cathode particles are constantly in motion and are suspended as they are agitated by the rotary bed system. In contrast, if artificial CEI coatings are formed on powders that are sitting on a substrate or in a crucible, the coating will form pinholes at the contact points, which will serve as hot spots for electrolyte decomposition.

A Schematic of ALD equipped with a rotary-bed attachment, ALD process for depositing a 5 nm thick amorphous Nb2O5 coating on SC-NMC particles, and composite cathode assembly. B High-resolution TEM micrograph showing amorphous Nb2O5 coating and layered (R-3m) structure of an SC-NMC particle. FFTs from marked regions are also presented. C HDAAF-STEM EDS maps showing conformal Nb2O5 coating and distribution of elements in an SC-NMC particle. XPS core scans corresponding to (D) Nb 3d peak (E) O 1s peak from Nb2O5-coated SC-NMC powder.

In this study, amorphous Nb2O5 films were deposited at a low temperature (175 ?C), without any high-temperature post-annealing step. In contrast, most of the available literature on Nb-based coatings and doping in layered oxide cathodes33,34,35,36,48,49 have reported that the development of Nb-based coatings typically entails a high-temperature (>=400 ?C) annealing process, which may lead to Nb doping in addition to the formation of a crystalline Nb-based coating. The extent and depth of Nb doping may further increase with increasing calcination temperature. Furthermore, high-temperature annealing often results in lithiation of NbOx surface films to form crystalline LiNbO3.

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