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β-NMR Measurements of $^{8}Li^{+}$ Motion in Ultra-Thin Al_2O_3 Capping Layers on NMC811 Cathode Materials

$LiNi_{0.8}Mn_{0.1}Co_{0.1}O_2$, commonly known as NMC811, is widely used as a cathode material in Li-ion batteries for electric vehicles due to its high energy density. Despite this, NMC811 and other Ni-rich layered cathode materials suffer from poor cycle life when compared to their lower Ni-containing analogues. This is caused by mechanical stress induced by anisotropic evolution of the crystal structure during Li extraction/insertion, which leads to microcracking and eventual disintegration of the cathode particles. Coating the particles with a protective layer such as Al_2O_3 can improve the long-term stability during repeated cycling, but little is known about how the lithium ions migrate through the protective coating. We report β-NMR measurements on $^{8}Li^{+}$ implanted in ultra-thin (12 and 30 nm thick) Al_2O_3 coatings deposited on two different orientations of NMC811. The results show that the local environment and motion of lithium ions is very different in the thin films compared with bulk Al_2O_3 and independent of the orientation of the NMC811 layer. The temperature dependence of the spin-lattice relaxation times indicates that $^{8}Li^{+}$ is hopping with an activation energy of 16(1) meV in the 30 nm thick Al_2O_3 capping layer and 8.8(6) meV in the 12 nm thick capping layer. These low-energy diffusion pathways in ultra-thin Al_2O_3 films allow it to act as protective coating and yet not impede the mobility of the lithium ions.

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