

Topotactic oxidation of Ruddlesden-Popper nickelates

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Layered perovskites — including the Dion–Jacobson, Ruddlesden–Popper, and Aurivillius families— exhibit a wide range of correlated electron phenomena, from high-temperature superconductivity to multiferroicity. Here, we report a new family of layered perovskites realized through topotactic oxidation of $\text{La}_{n+1}\text{Ni}_n\text{O}_{3n+1+\delta}$ ($n = 1 - 4$) Ruddlesden–Popper nickelate thin films. Post-growth ozone annealing induces a substantial c-axis expansion — 17.8% for $\text{La}_2\text{NiO}_{4+\delta}$ ($n = 1$) — that monotonically decreases with increasing n . Surface X-ray diffraction and Coherent Bragg Rod Analysis reveal that this expansion arises from the intercalation of approximately 0.7 oxygen atoms per formula into the rock salt spacer layers, far exceeding the previous record of $\delta \sim 0.3$ for any Ruddlesden–Popper oxide. These oxygen-intercalated phases form a new structural class, with a spacer layer composition intermediate between the Ruddlesden–Popper and Aurivillius phases. Oxygen intercalation induces metallicity, suppresses oxygen octahedral rotations, and enhances nickel-oxygen hybridization — features associated with high-temperature superconductivity. Our work establishes topotactic oxidation as a powerful approach to accessing highly oxidized, metastable phases across a broad range of layered oxide systems, offering new platforms to engineer electronic properties via intercalation chemistry