

Figure S1: Observed, calculated and difference plots for the 'single phase' fit to the neutron powder diffraction data. The expanded region shows the poor fit in the region $45<2\theta/^{\circ}<90$. wRp = 3.86%, Rp = 3.00%, χ^2 = 3.222. Refined lattice parameters of LaNiO₂ phase: a = 3.9610(9) Å, c = 3.3816(2) Å. Refined lattice parameter of nickel impurity : a = 3.5255(4) Å.

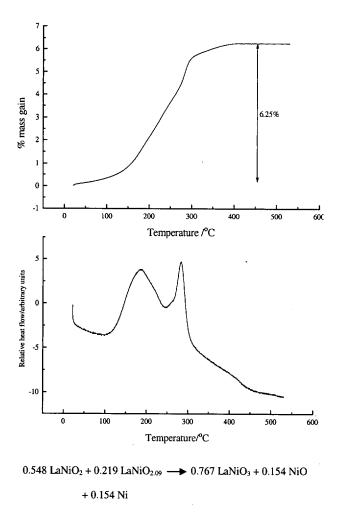


Figure S2: TGA and DSC data showing the reoxidation reaction of LaNiO_{2.03} under O₂. 0.077 La₂O₃ has been omitted from both sides of the equation for clarity. The La₂O₃ is required to satisfy elemental analyses showing the La:Ni ratio of the sample is 1:1, despite no diffraction features assignable to La₂O₃ being observed. (There is no evidence in the Rietveld profile refinement that the LaNiO_{2+x} phases are nickel deficient.)

Phase 1	
Strain parallel to [001]	0.89(4)%
Strain perpendicular to [001]	0.87(2)%
particle size parallel to [001]	78(8)Å
particle size perpendicular to [001]	591(20)Å
Phase 2	
Strain parallel to [001]	2.3(4)%
Strain perpendicular to [001]	0.5(1)%
Particle size parallel to [001]	79(7)Å
Particle size perpendicular to [001]	536(26)Å
Nickel metal	
Strain	1.9(2)%
Particle size	513(90)Å

Table S1: Strain and particle size values evaluated from refined profile coefficients of the majority LaNiO_2 and minority LaNiO_{2+x} phases.