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Figure S1. APXPS survey spectra taken, from bottom to top, under 100 mTorr O<sub>2</sub> at 250°C, 100 mTorr H<sub>2</sub> at 250°C, 300 mTorr CO+H<sub>2</sub>(1:2) at 225°C, 100 mTorr H<sub>2</sub> at 350°C, 300 mTorr CO+H<sub>2</sub>(1:2) at 225°C, 100 mTorr H<sub>2</sub> at 480°C and 300 mTorr CO+H<sub>2</sub>(1:2) at 225°C.



Figure S2. XAS spectra of various Mg and Co references, and CoO/MgO nano-composite, comparing local (STXM) and bulk ensembles (XAS), at (a) Mg K and (b) Co  $L_3$  absorption edges.



Figure S3. (a) Mg 2p+Co 3p XPS spectra are shown under various conditions. (b) Plots of fractions of Mg and Co at 350 eV photon energy. (c) Plots of full width at half maxima of Mg 2p peak at two different photon energies. XPS spectra in (a) were acquired progressively from bottom to top, and numbered as in (b) and (c).



Figure S4. (a) C 1s APXPS spectra acquired using 630 eV photons and under various conditions. (b) Plots for fractions of adsorbed CO ( $CO_{(ads)}$ , red), lattice carbonate ( $CO_3^{2-}$ , blue) and sp<sup>3</sup>-like surface carbon (black). XPS spectra in (a) were acquired progressively from bottom to top, and numbered as in (b).