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Supporting Information

The role of ions and reaction sites for electrochemical reversible charge cycling in layered nickel hydroxides

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S1: Cycling of sputtered NiO film in KOH and Li-PC

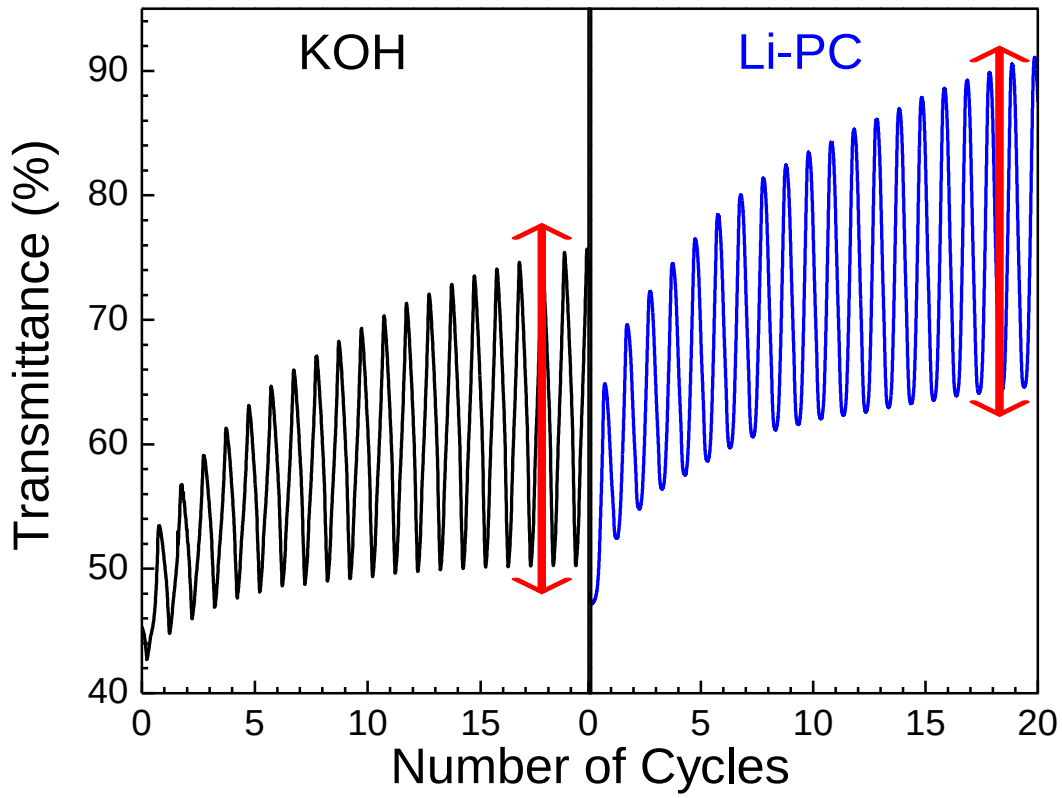

Fig. S1 Transmittance variation of defective sputtered NiO cycled in KOH and Li-PC

The sputtering of NiO is achieved using unbalanced magnetron sputtering of a Ni metal target at radio frequency. The sputtering power is 300W and the pressure is controlled at 20mTorr, with a Ar:O_2 ratio of 5:2 in the sputtering gas mixture, to achieve a largely defective film that is shown by its low transmittance of <50%. Figure S1 shows that after about 20 cycles, the modulation in KOH and Li-PC is ~30% as indicated by the red arrow. The similar achievable modulation in both KOH and Li-PC is in great contrast to that achievable for the $Ni(OH)_2$ sample and shows the importance of defects as reactive sites.

S2: SIMS depth profile

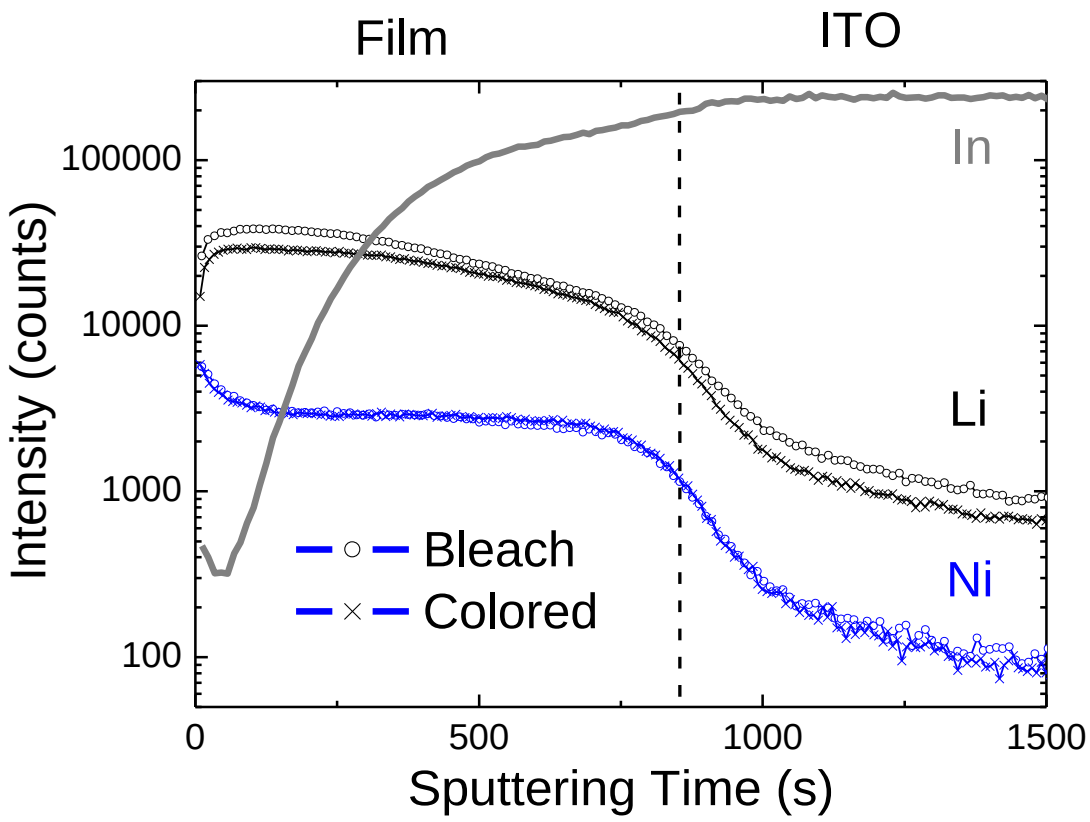

Fig. S2 Depth profile SIMS for bleach and colored sample in Li-PC

The same hydrated NiOOH sample in the bleach and colored state is dissected for probing with SIMS depth profiling. The plot is normalized with the Ni signal. Porous nature of the film is shown with a less sharp profile of the In but the interface can still be identified from the flattening out of the profile. The depth profile shows the cycling out of Li during coloring that was expected. However, since we started with a NiOOH sample, the colored film should be void of Li profile. The presence of Li remaining in the film for the colored sample shows the possible formation of Li-OH that remains trapped.