

SUPPORTING INFORMATION

***Operando* XRD and electrogravimetry coupling to analyze species transfers during redox processes in Ni/Fe layered double hydroxide**

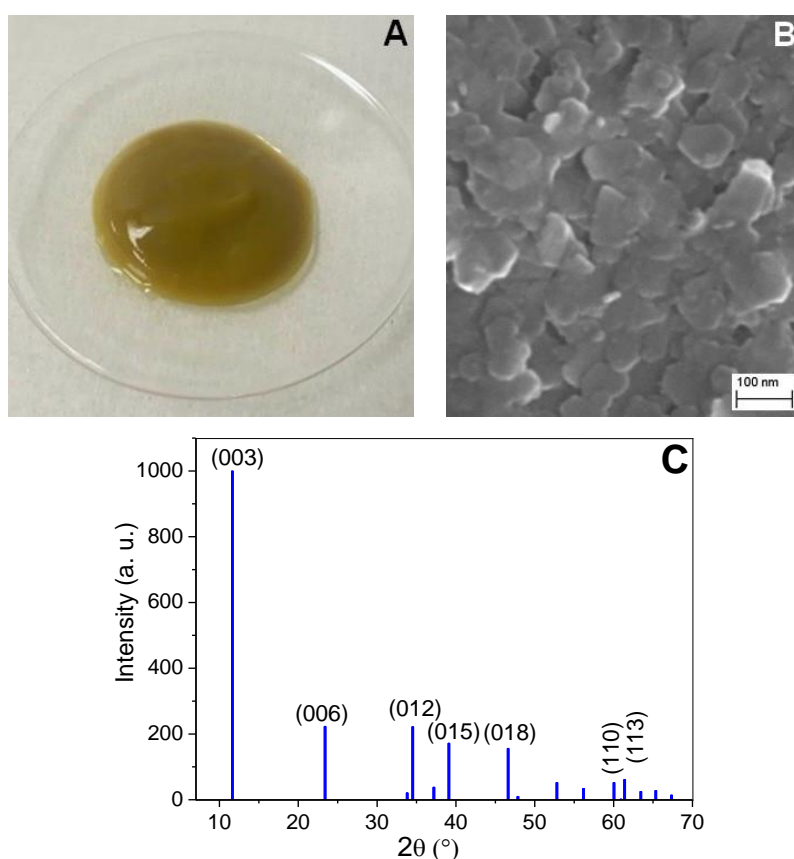
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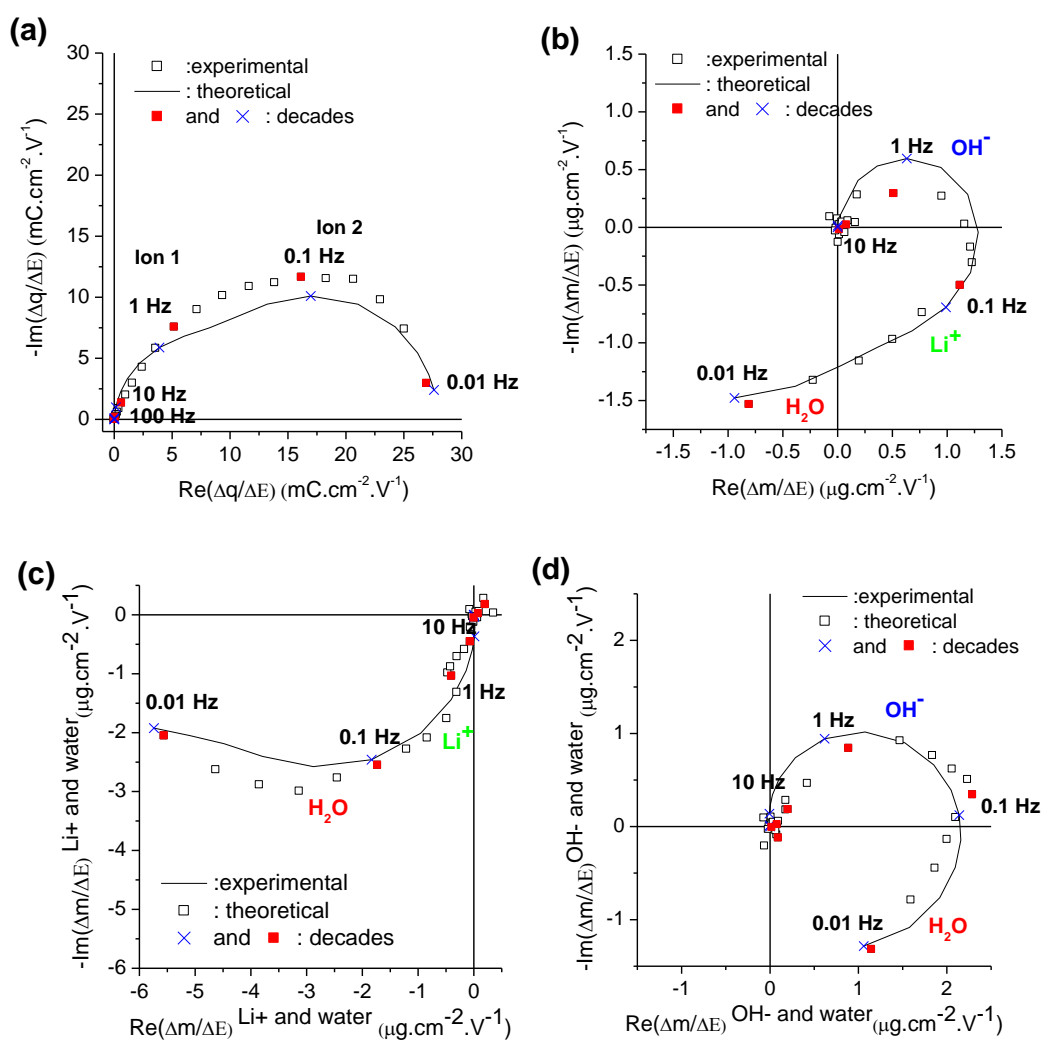
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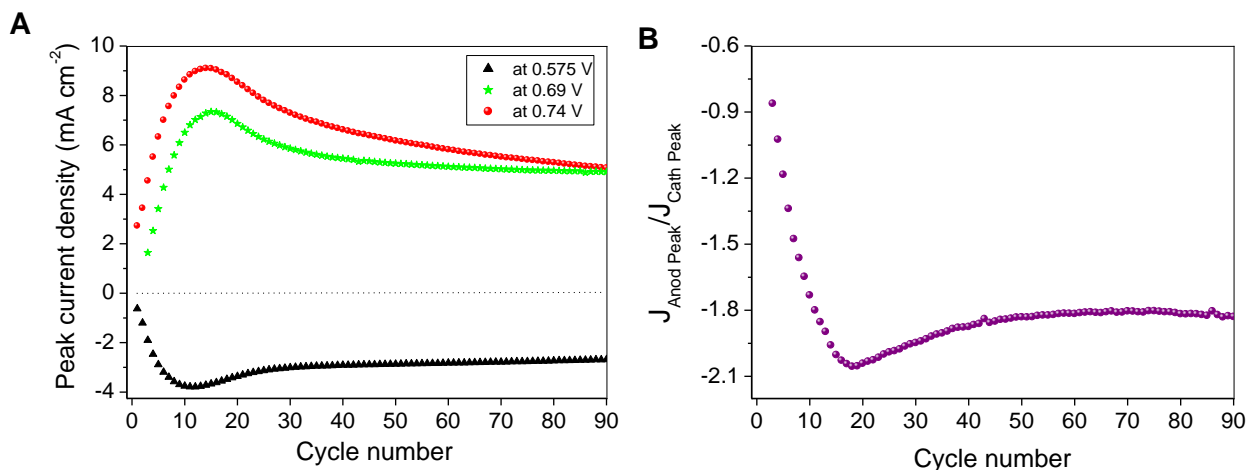
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**Figure S1.** A) Photo of the Ni/Fe-LDH slurry. B) SEM micrograph of Ni/Fe-LDH platelets deposited on a substrate by drop-casting. C) XRD pattern of  $\text{Ni}_2\text{Fe}_6(\text{OH})_{16}(\text{CO}_3)(\text{H}_2\text{O})_{4.5}$ , data are from ICDD card n°01-082-8040.



**Figure S2.** Fitting of the experimental data of a thin film of Ni/Fe-LDH immersed in 1 M LiOH at 0.65 V/SHE: (a)  $\Delta q/\Delta E$  TF, (b)  $\Delta m/\Delta E$  transfer function, (c)  $\Delta m/\Delta E$  ( $\text{Li}^+$  +  $\text{H}_2\text{O}$ ) partial transfer function (after removing the contribution of  $\text{OH}^-$ ), (d)  $\Delta m/\Delta E$  ( $\text{OH}^-$  +  $\text{H}_2\text{O}$ ) partial transfer function (after removing the contribution of  $\text{Li}^+$ ).



**Figure S3.** Ni/Fe-LDH in LiOH. (A) Peak current densities at each cycle: Ni(II) oxidation in green, Ni(III) reduction in black and anodic limit due to OER in red. (B) Ratio of the intensity of the Ni(II) oxidation peak over the intensity of the Ni(III) reduction peak vs. cycle number, in 1 M LiOH at  $10 \text{ mV}\cdot\text{s}^{-1}$  (data from Figure 4).