Supplementary Information

Alkoxide-intercalated CoFe-layered double hydroxides as precursors of colloidal nanosheet suspensions: structural, magnetic and electrochemical properties

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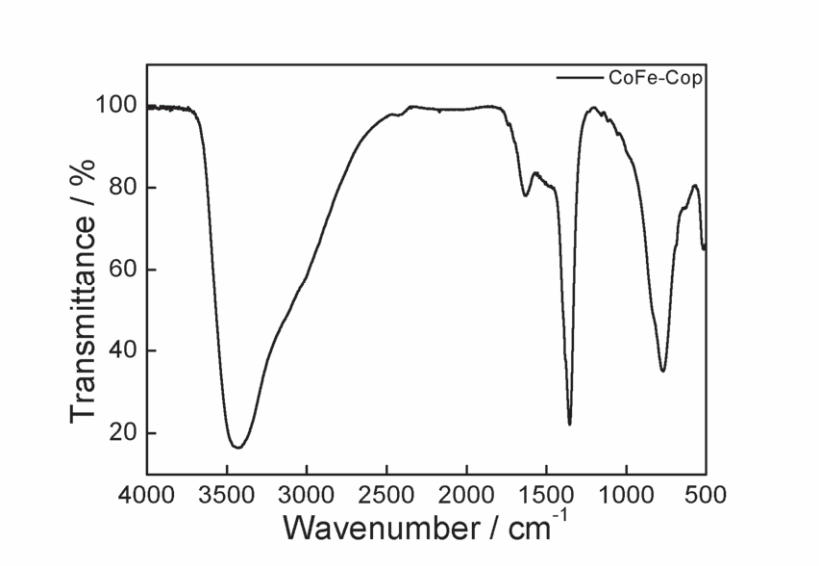
Contents

SI 1: Physical Characterization of CoFe-LDH synthesized *via* coprecipitation route.

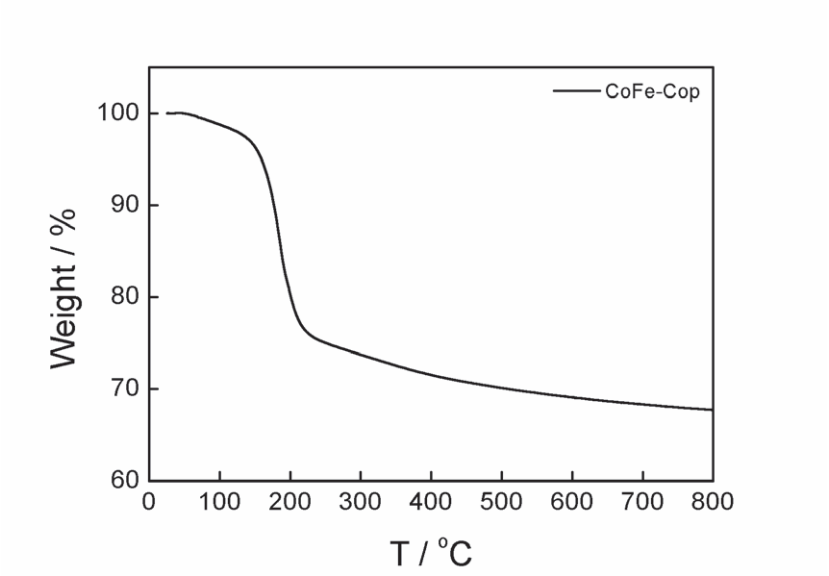
SI 1.I: Chemical Composition.

Metallic atomic composition of bulk samples was determined by means of electron probe microanalysis (EPMA) performed in a Philips SEM-XL30 equipped with an EDAX microprobe. Carbon, hydrogen and nitrogen contents were determined by microanalytical procedures using an EA 1110 CHNS-O elemental analyzer from CE Instruments. The proposed chemical formula was estimated to be: $[Co_{0.67}Fe_{0.33}(OH)_2](CO_3)_{0.16}\cdot 0.2H_2O$; Co/Fe = 2 / 1. (C,H,N, calc: 1.9, 2.3, 0; found: 1.56, 2.44, 0.14).

SI 1.II: FT-IR Spectra.

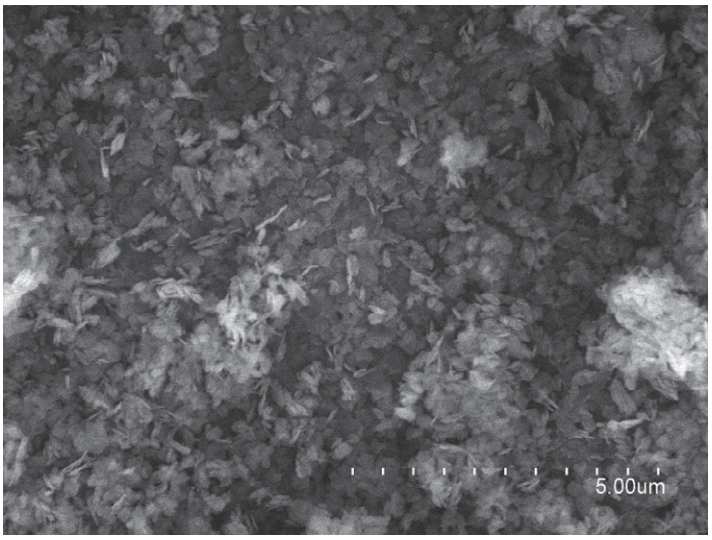


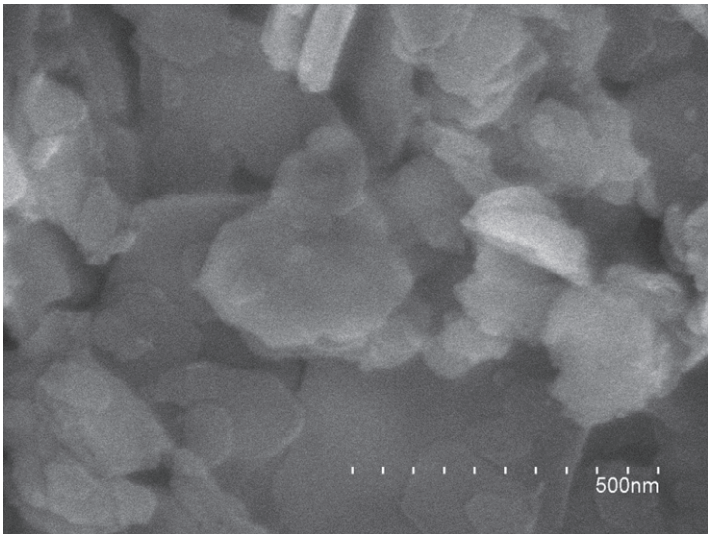
SI 1.III: Thermogravimetric Analysis.



SI 1.IV: FESEM images of CoFe-LDH Cop.

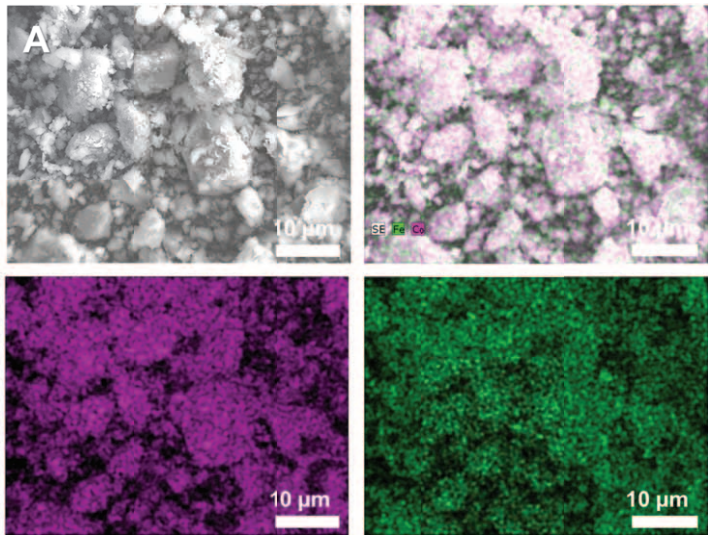
FESEM images of the CoFe-LDH Cop sample show a wide distribution of sizes of several hundred nanometers.

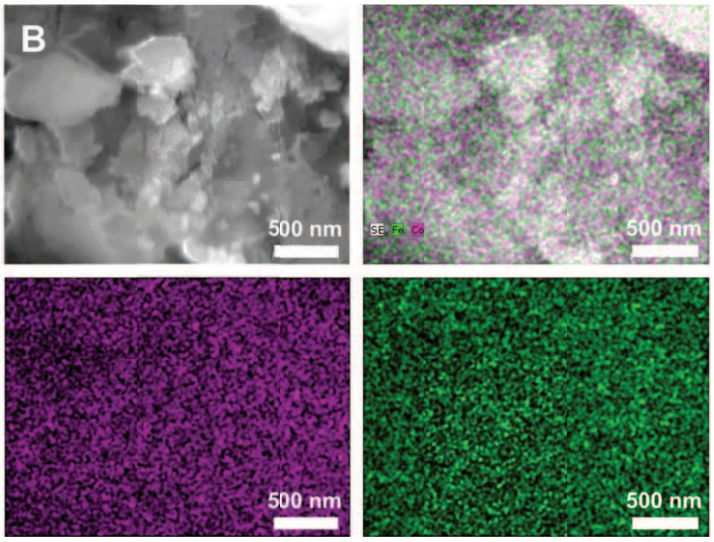




SI 2. FESEM-EDS study of CoFe-LDH.

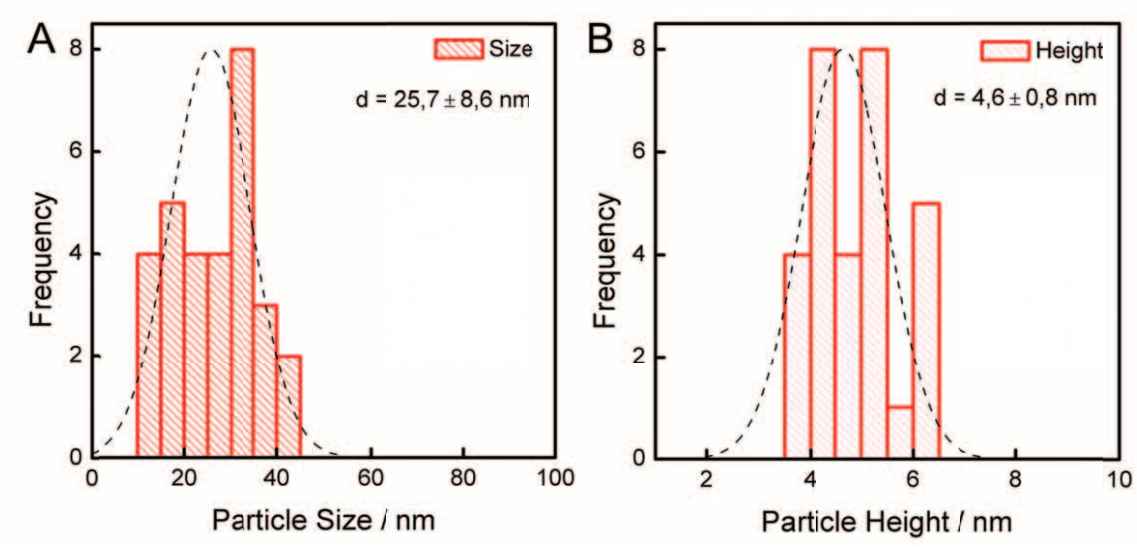
(A and B) Field-emission scanning electron microscopy with elemental mapping (FESEM-EDS) images of CoFe-LDH measured at different magnifications can be observed in (A) and (B), showing the homogeneous distribution of Co (purple) and Fe (green) in the sample, discarding the presence of segregated phases.



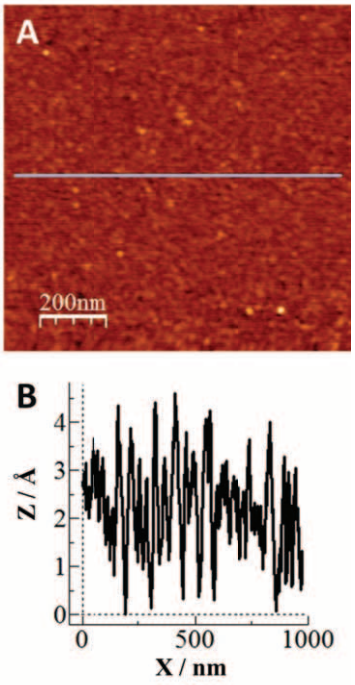


SI 3. AFM study of exfoliated CoFe-LDH.

In order to further determine the lateral dimensions and height of our LDH nanosheets we have measured several exfoliated CoFe-LDHs. The following Figure shows the corresponding AFM histograms of the exfoliated particles in water, depicting the (A) particle-size data and (B) the corresponding particle-height.

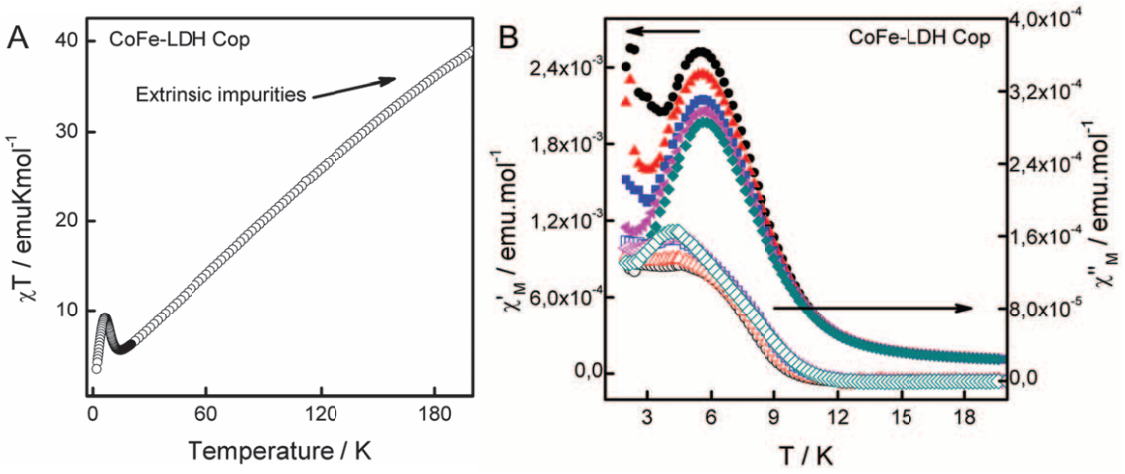


Control experiment on SiO_2 prepared by the same procedure with its height profile.



SI 4. Magnetic properties of CoFe-LDH synthesized *via* coprecipitation route.

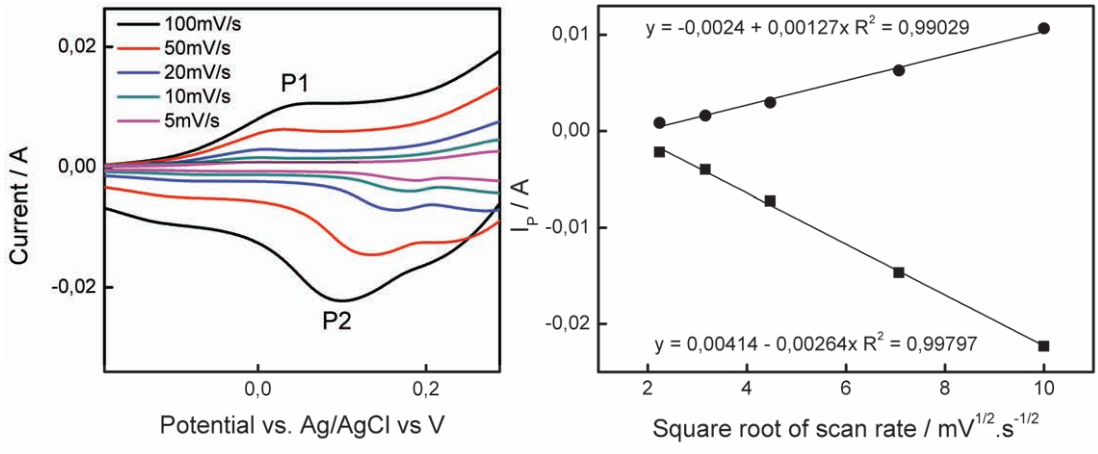
(A) The temperature-dependent magnetic susceptibility, χ, reported as, χT' versus T shows an almost linear increase above ca. 25 K for the CoFe-LDH Cop sample, whilst no increase was observed for the CoFe-LDH sample (Figure 4 main text). In both cases (CoFe-LDH and CoFe-LDH Cop), the drop observed below 20 K defining a maximum corresponds to the intrinsic cooperative magnetism in the LDH layers. (B) The measurements of the ac susceptibility revealed a temperature for the onset of the spontaneous magnetization (TM) of ca. 11 K, 4 K higher that that exhibited by the pure CoFe-LDH (Table 1 main text), indicative of size effects, with an average size of several hundred nanometers for the CoFe-LDH Cop platelets in contrast with the homogeneous distribution of sizes for the CoFe-LDH, as depicted in the FESEM measurements.



SI 5: Diffusional behaviour of CoFe-LDH: peak current (i_p) *vs.* square root of scan rate ($\nu^{1/2}$) plots.

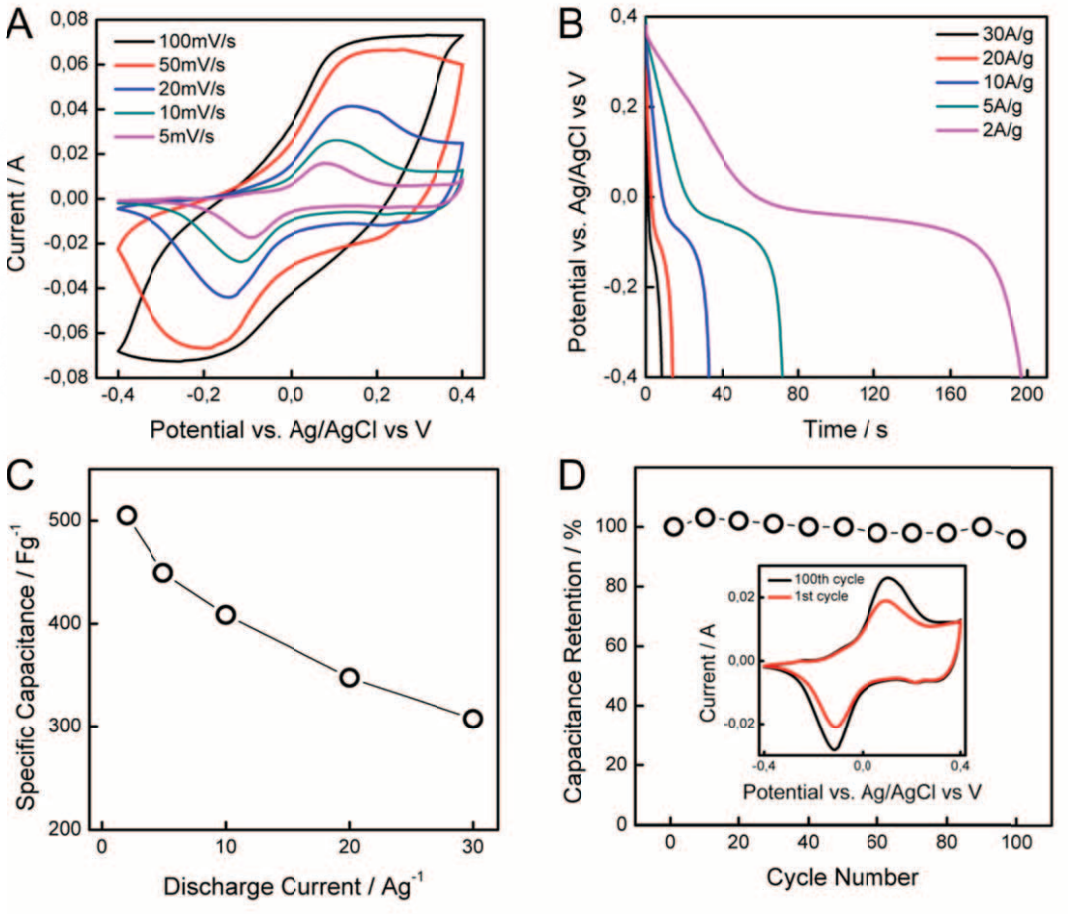
Cyclic voltammograms of CoFe-LDH at different scan rates (left) indicating the fitted peaks.

Plot of the peak current *vs*. square root of scan rate ($\nu^{1/2}$) for peaks P1 and P2 (right).



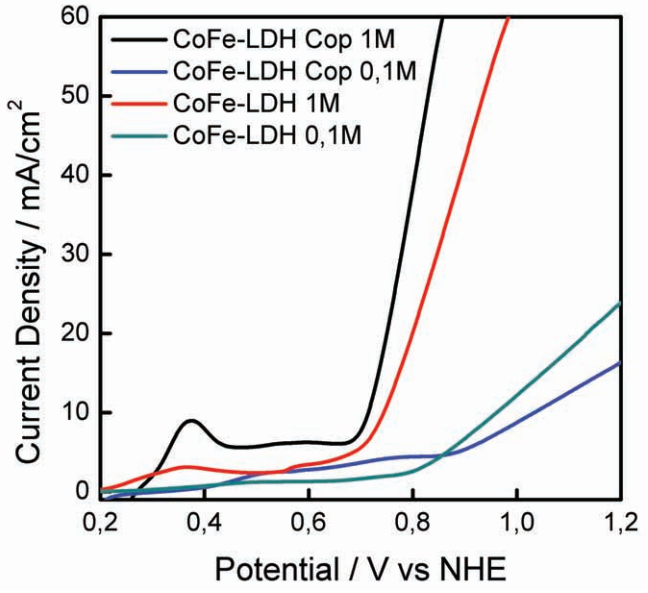
SI 6: Electrochemical properties of CoFe-LDH Cop

(A) CV curves at various scan rates in 6 M KOH aqueous solution. (B) Galvanostatic discharge curves and (C) specific capacitance of the material at different discharge current densities. (D) Specific capacitance *vs*. cycle number at a current density of 40 Ag^{-1}.



SI 7. Comparison of LSV curves for CoFe-LDH and CoFe-LDH Cop in 0.1 M and 1 M KOH.

The direct comparison between CoFe-LDH and CoFe-LDH Cop leads to a similar behaviour strongly dependent on pH, with some differences probably arising from the extrinsic sources as well as the different particle size and/or morphology. Further studies are needed in order to clarify these differences on the electrocatalytic behaviour.



SI 8: Linear sweep voltammetry curves of CoFe-LDH.

LSV curves obtained with freshly prepared CoFe-LDH and 10000 s after use under chronoamperometric measurements at 0.83 V (0.1 M) and 0.75 V (1 M) vs ENH.