Supplementary Information

One-Step Direct Synthesis of Layered Double Hydroxide Single Layer Nanosheets

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Preparation Method

**Materials.** Formamide (99%, Alfa Aesar), N,N-dimethyl formamide (≥99.8%, Sigma-Aldrich), Mg(NO$_3$)$_2$·6H$_2$O (98%, Alfa Aesar), Co(NO$_3$)$_2$·6H$_2$O (>98%, Alfa Aesar), Al(NO$_3$)$_3$·9H$_2$O (99%, Acros), sodium hydroxide (98%, Macron), and NaNO$_3$ (>98%, Alfa Aesar) were all used as received without further purification.

**Characterization.** X-ray diffraction (XRD) patterns were recorded on a Bruker D8 diffractometer with Bragg-Brentano θ-2θ geometry (40 kV and 30 mA), using a graphite monochromator with Cu Ka radiation (λ = 1.542 Å). For solution and gel-like samples, they were cast as a thin film on a silicon wafer, which was covered by a Mylar® film during the characterization. In this way, the liquid and gel-like sample can maintain a flat surface during characterization. In addition, the synthesized LDH nanosheets were cast on a clean silicon wafer and dried at room temperature for XRD characterization.

Transmission electron microscopy (TEM) imaging was carried out using a FEI Tecnai G2 F20 with field emission gun (FEG) at a working voltage of 200 kV. Observations were made through the holes of the carbon support film, so that no noise from the support film was introduced.

Atomic force microscopy (AFM) characterization was performed on an Asylum Research MFP-3d AFM. AFM images were obtained under the tapping mode using a silicon tip coated with chromium/gold with a force constant of 40 N/m. LDH nanosheet samples were diluted into ca. 0.01 mM and drop-coated on a clean silicon wafer for AFM imaging.

**Direct synthesis of colloidal single layer nanosheets.** The traditional titration method was slightly modified to directly synthesize single layer MgAl-LDH by adding 23 vol% formamide as an inhibitor. A 10.0 mL solution composed of Mg(NO$_3$)$_2$ (0.040 M) and Al(NO$_3$)$_3$ (0.010 M) was added drop by drop to a solution of 10.0 mL NaNO$_3$ (0.010 M) containing 23 vol% formamide. Simultaneously, a solution of 0.25 M NaOH was added dropwise under magnetic stirring at 80 °C to maintain the system at a pH value of ca. 10. The reaction was completed within 10 min. After that, the prepared sample was centrifuged and washed with water. By repeating the process three times, single layer LDH nanosheets
dispersed in water were obtained. A control sample of conventional layered MgAl-LDH was prepared using the same formulation and under the same conditions but in the absence of formamide.

MgAl-LDH nanosheets were also synthesized using 23 vol% $N$, $N$-dimethyl formamide as an inhibitor through the same procedures.

Similarly, Co$^{2+}$/Al$^{3+}$ layered double hydroxide (CoAl-LDH) was also prepared [using Co(NO$_3$)$_2$ to replace Mg(NO$_3$)$_2$] following the same method in the presence of formamide. In the synthesis of CoAl-LDH nanosheets, 20.0 mL solution of Co(NO$_3$)$_2$ (0.032 M) and Al(NO$_3$)$_3$ (0.018 M) was added drop by drop into a solution of 20.0 mL NaNO$_3$ (0.018 M) containing 23 vol% formamide. The reaction was carried out at 80 °C with magnetic stirring, during which a solution of 0.25 M NaOH was added dropwise to maintain the system at a pH value of ca. 10.
Figure S1. Structure of layered double hydroxide.
Figure S2. EDX spectrum of MgAl-LDH nanosheets prepared in the presence of formamide.
Figure S3. XRD patterns of the pre-synthesized MgAl-LDH before (insert, dry solid) and after being stirred in an aqueous dispersion containing 23 vol% formamide at 80 °C for 10 min.
Figure S4. XRD patterns of the gel like MgAl-LDH nanosheets collected after centrifuge.
Figure S5. SEM image of the control MgAl-LDH sample synthesized in the absence of formamide.
Figure S6. XRD patterns of: (I) aqueous dispersion of CoAl-LDH control sample synthesized in the absence of formamide; (II) aqueous dispersion of directly synthesized CoAl-LDH single layer nanosheets in the presence of formamide; and (III) re-stacked CoAl-LDH nanosheets on a silicon wafer after drying. Inset: XRD pattern of CoAl-LDH control sample in powder form.
Figure S7. XRD patterns of: (I) aqueous dispersion of MgAl-LDH control sample synthesized in the absence of $N,N$-dimethyl formamide; (II) aqueous dispersion of directly synthesized MgAl-LDH single layer nanosheets in the presence of $N,N$-dimethyl formamide; and (III) re-stacked MgAl-LDH nanosheets on a silicon wafer after drying.
References:
