Supporting information for:

Direct Synthesis of Unilamellar MgAl-LDH Nanosheets: Stacking in Aqueous Solution

Dujuan Liang, Wenbo Yue, Genban Sun, Dong Zheng, Kenta Ooi and Xiaojing Yang*

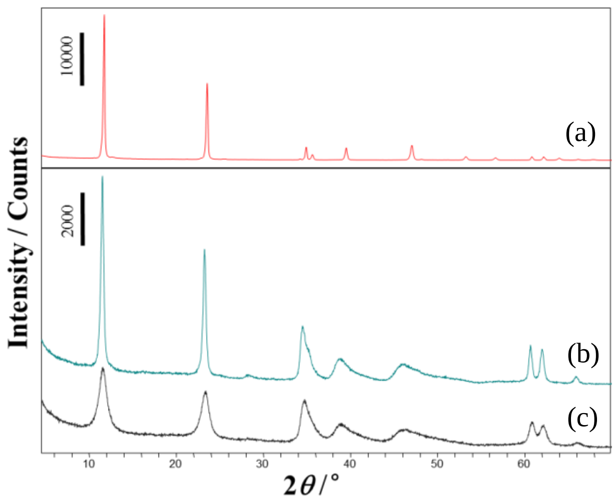

Figure S1. XRD pattern of MgAl-LDH for (a) synthesized by homogenous precipitation method,[25] (b) Na_2CO_3-and (c) water-washed sample.

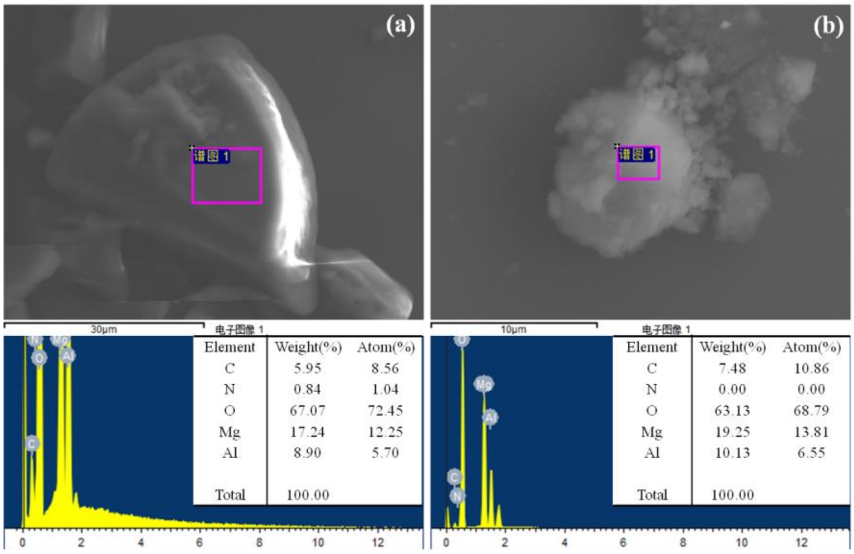

Figure S2. EDX result of (a) water- and (b) Na_2CO_3-washed samples. The Mg/Al ratios are close to 2.

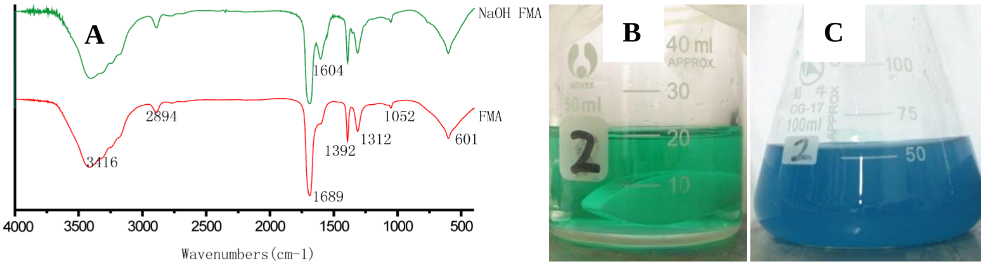

Figure S3. (A) FT-IR spectra for formamide (FMA) and the product obtained after FMA mixed with NaOH for 30 min. The 1604cm^{-1} band of C=O in -COO^{-} indicate the $HCOO^{-}$ generation; (B) before and (C) after add NaOH-formamide solution to green Ni^{2+}, blue $[Ni(NH_3)_6]^{2+}$ can prove the large amount of NH_3 existence.

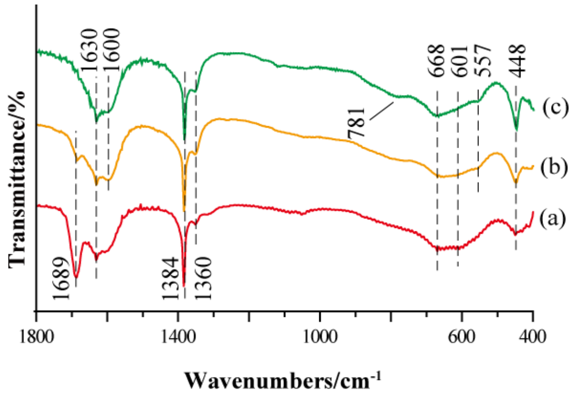

Figure S4. FT-IR spectra of as-prepared R2.0 sample after washed by water 1(a), 3 (b) and 5 (c) times, exhibiting the removal process of formamide during washing the as-prepared sample using decarbonized water, which is characterized by the disappearance of the bands at 1689 and 601 cm^{-1}.

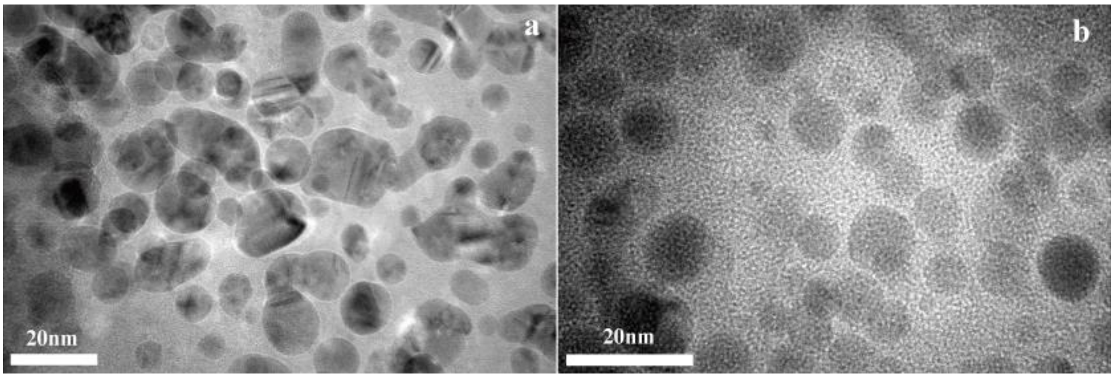

Figure S5. TEM images for nanosheets (a) before and (b) after removing extra salts and hydrothermal aging. Chen et al.[32] pointed out that extra metal cation and inorganic anions can cause LDH nanosheets' aggregation effectively in aqueous solution. The remove-extra-salts method of redispersing the centrifuged original gel to pure formamide can facilitate evenness in diameter and form regular round shape through Brownian motion and Ostwald ripening.

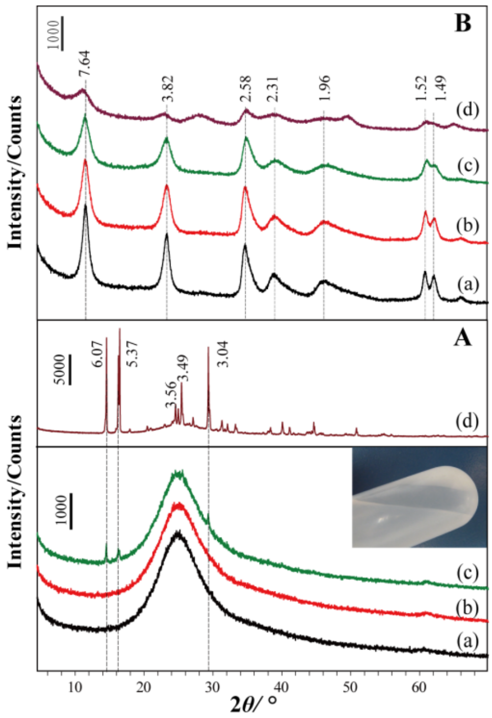

Figure S6. XRD patterns in (A) wet-state with FMA and (B) dry-state after Na_2CO_3(aq) washing, (a) (b) (c) and (d) corresponding to room temperature (RT), 60, 80 and 100℃ aging temperature for 1 day. The inset is the photo of 80℃-sample after centrifugation. As temperature increases, unidentified crystallites appear. After washing with Na_2CO_3 colloid nanosheets can stack to sharper diffraction peaks, while crystallite cannot.

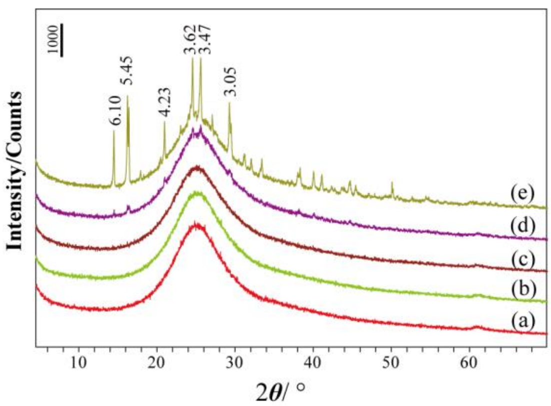

Figure S7. XRD patterns of the samples for different time at 60℃, (a) (b) (c) (d) (e) corresponding to the aging time of 1, 2, 3, 4, 5 days. Long-time heating could get analogous result with high-temperature heating (Figure S4). Excessive temperature and time will decrease the stability of 2D sheets.

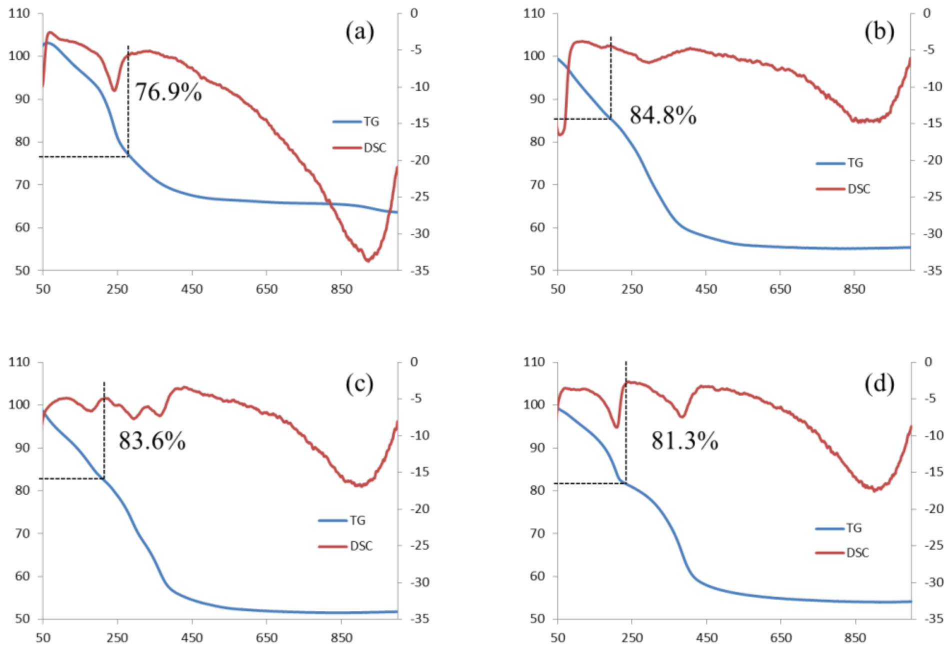

Figure S8. The TG-DSC curves of R0.8 (a), R1.0 (b) R2.0 (c) water-washed samples and R2.0 Na_2CO_3-washed sample (d). The weight losses around 250 ℃ corresponding to the loss of interlayered water with endothermic peaks. The different intercalated species lead to the different weight loss temperatures and thermal analysis.

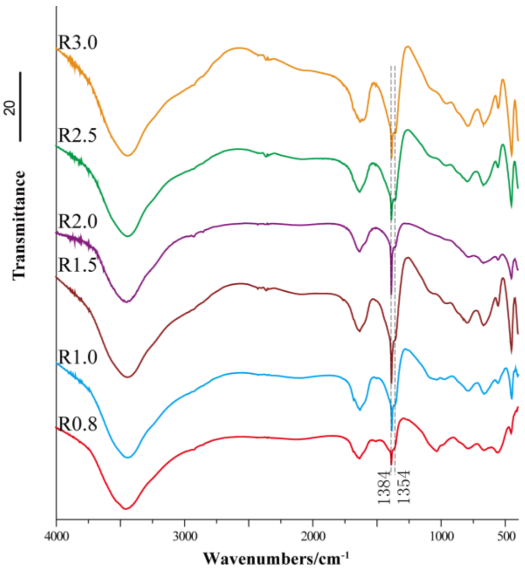

Figure S9. The FT-IR spectra for water-washed samples. Both the bands of nitrate (1384 [ν(NO_3^{-})]) and carbonate (1360 (ν_{C-O}) and 781 (δ_{C-O}) cm^{-1}) exsit.

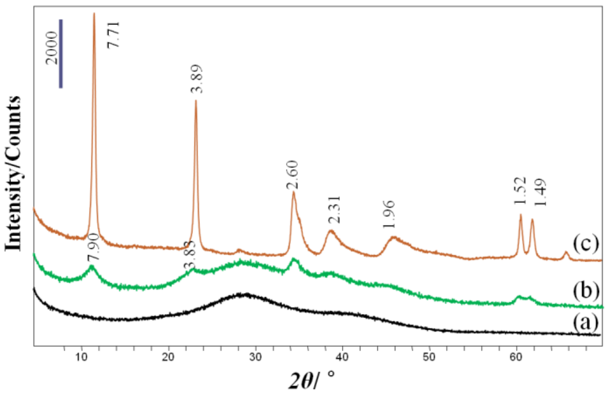

Figure S10. XRD patterns of synthetic $Al(OH)_3$ (a) and transferred MgAl-CO_3^{2-}-LDH under RT (b) and 100 ℃-20h hydrothermal treatment (c). we add alkaline solution (aq) of NaOH and CO_3^{2-} to a suspension (aq) of Mg^{2+} and $Al(OH)_3$ (Mg/Al=2) and results to a typical CO_3^{2-}-LDH XRD diffraction pattern.

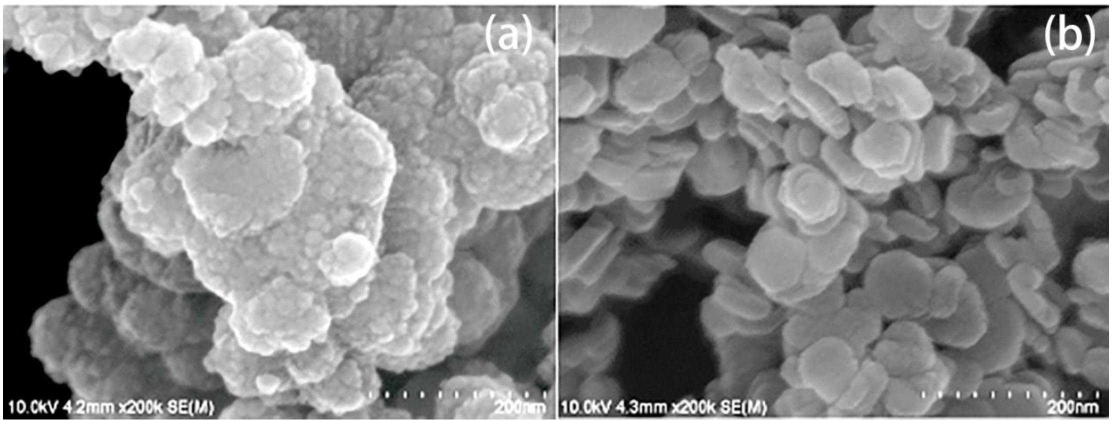

Figure S11. SEM images of $Al(OH)_3$ (a) and hydrothermal-treatment transferred hexagon shape $MgAl\text{-}CO_3^{2-}$-LDH for well growth in 3D (b). The formation of small LDH particles is based on $Al(OH)_3$ skeleton and then small particles grow up though Brownian motion and collision.

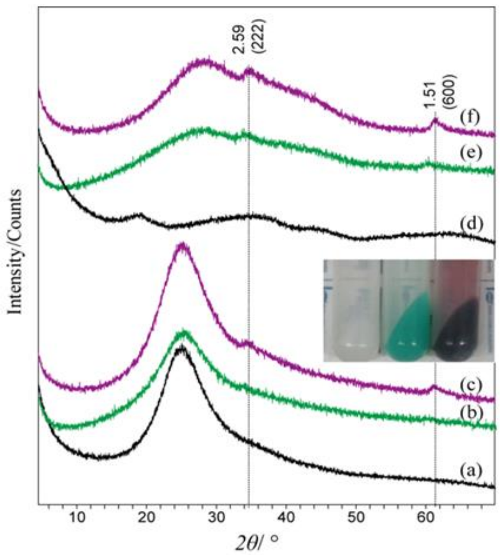

Figure S12. XRD patterns of ZnAl (a) (d), NiAl (b) (e), and CoAl (c) (f) systems synthesized by the same method as MgAl. (a)-(c) are colloids showing no clear 2D characteristic diffraction peak, and (e)-(f) are water-washed state showing little lamellas restack to diffract 3D peaks. The inset is colloids images of the three systems after centrifuging, and the colors are derived from $[Zn(NH_3)_4]^{2+}$, $[Ni(NH_3)_6]^{2+}$ and $[Co(NH_3)_6]^{2+}$ complex ions. $Zn(OH)_2$, $Ni(OH)_2$, $Co(OH)_2$ and $Al(OH)_3$ all are amphoteric.

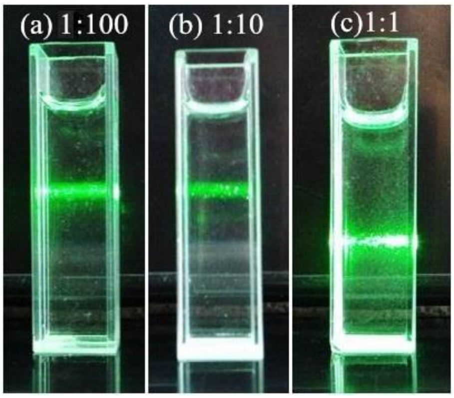

Figure S13. The Tyndall effects of different water: formamide (volume ratio) reagent to exfoliate MgAl-NO_3-LDH. Some crystal can be seen in the light path under ratio1:10, and the light path is discontinuous when the ratio up to 1:1.

Table S1. The Concentration of Mg and Al in Aqueous Solution during Transformation Process

c (mg/L)	RT for 5 seconds	RT for 1day	100 ℃ for 20 hours
Mg	0.368	0.295	0.329
Al[a]	—	—	—

[a]"—" means the tested data are below 0.001 mg/L

Table S2. The CHN Analyses of Some Samples

samples		N%	C%	H%
water-washed	R0.8	0.18	2.29	3.789
	R1.0	0.61	2.83	4.188
	R1.5	1.65	2.49	3.776
	R2.0	1.54	3.87	3.957
	R2.5	1.14	3.45	3.895
	R3.0	0.98	3.28	3.606
Na_2CO_3-washed	R2.0	0.01	3.10	4.088

Table S3. Mg/Al Ratios Analyzed for the Rx Samples after Different Wash Time

Mg/Al	R0.8	R1.0	R1.5	R2.0	R2.5	R3.0
Wash 30 min/time	0.12	0.39	1.18	1.68	1.64	1.62
Wash 5 min/time	0.42	1.03	1.90	1.92	1.89	1.98