Electronic Supporting Information

Large-Scale Synthesis of Highly Dispersed Layered Double Hydroxide Powders Containing Delaminated Single Layer Nanosheets

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1. Experimental details

1.1 Synthesis

A conventional sample of Zn_2Al-borate LDH was prepared by adding 100 ml $Zn(NO_3)_2 \cdot 6H_2O$ (0.075 mol) and $Al(NO_3)_3 \cdot 9H_2O$ (0.0375 mol) solution drop-wise

into a 100 ml H_3BO_3 (0.187 mol) solution. The pH of the precipitation solution was controlled at *ca*. 8.3 using a NaOH (1 M) solution. During the synthesis, the system was purged with N_2 gas to prevent the contamination by atmospheric CO_2. The mixture was aged at 65^{o}C for overnight. The obtained white solid were washed with H_2O until pH was close to 7 and dried at 65^{o}C overnight.

The new **AMOST** Zn_2Al-borate LDH was prepared as described above except the water washed LDH slurry was re-dispersed in acetone and stirred at room temperature for 1 h. After that, the LDH was filtered and washed thoroughly with acetone again. Finally the product was dried at 65^{o}C overnight. Similarly, Mg_3Al-borate LDH was synthesized at pH 9 using 100 ml $Mg(NO_3)_2 \cdot 6H_2O$ (0.075 mol) and $Al(NO_3)_3 \cdot 9H_2O$ (0.025 mol) solution drop-wise into a 100 ml H_3BO_3 (0.187 mol) solution followed by the new **AMOST** method described above.

1.2 Characterization

X-ray diffraction (XRD) patterns were recorded on a PANalytical X'Pert Pro instrument in reflection mode with Cu K□ radiation. The accelerating voltage was set at 40 kV with 40 mA current (λ = 1.542A°) at 0.01^o s^{-1} from 1^o to 70^o with a slit size of 1/4 degree. Transmission Electron Microscopy (TEM) analysis was performed on JEOL 2100 microscope with an accelerating voltage of 400 kV. Samples were dispersed in ethanol with sonication and then cast onto copper TEM grids coated with lacey carbon film. Scanning Electron Microscopy (SEM) and Energy dispersive X-ray Spectrometry (EDS) analyses were performed on a JEOL JSM 6100 scanning microscope with an accelerating voltage of 20 kV. Powder samples were spread on

carbon tape adhered to an SEM stage. Before observation, the samples were sputter coated with a thin Platinum layer to prevent charging and to improve the image quality. BET specific surface areas were measured from the N_2 adsorption and desorption isotherms at 77 K collected from a Quantachrome Autosorb-6B surface area and pore size analyzer. Before each measurement, LDH samples were first degassed overnight at 110^{o}C. The apparent density was determined by a method described as follows. LDH dry powder was filled into a 2 ml disposable tube, and the solid was packed as tight as possible by manually tapping(vibrating) for 2 minutes. The weight of the disposable tube was measured before and after the packing, and the weight of such packed 2 ml LDH powder was obtained from the weight difference. The apparent density of each LDH was calculated using the following equation:

Apparent density = LDH weight (g)/LDH volume (2 ml)

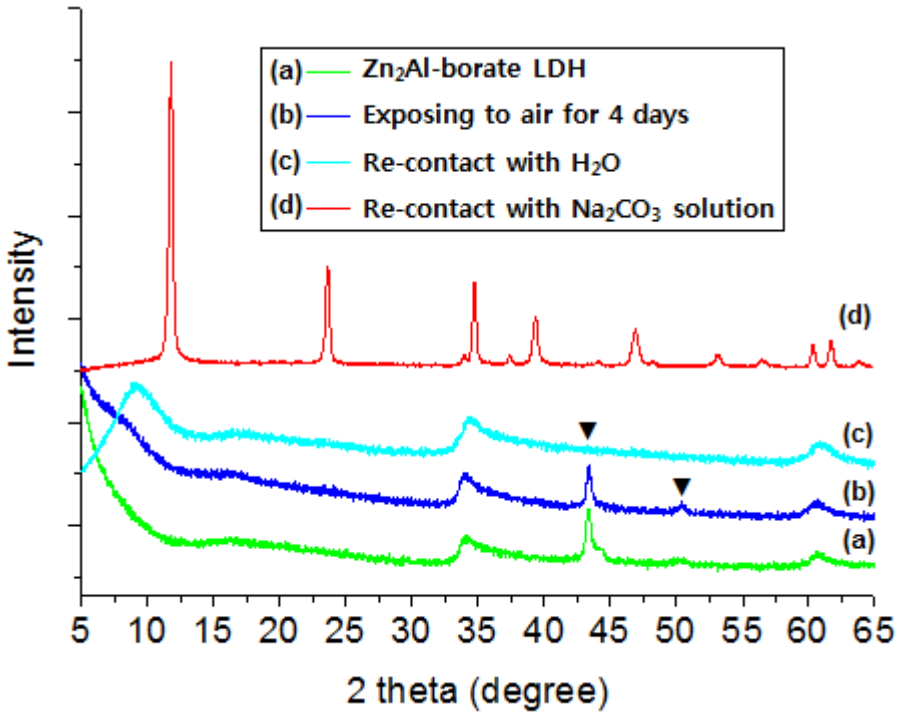

Figure S1. XRD patterns of acetone washed Zn_2Al-borate LDH (a) dried powder, (b) exposed to air for 4 days, (c) re-contacted with H_2O, and (d) re-contact with Na_2CO_3 solution. (▼) reflections from the sample holder.

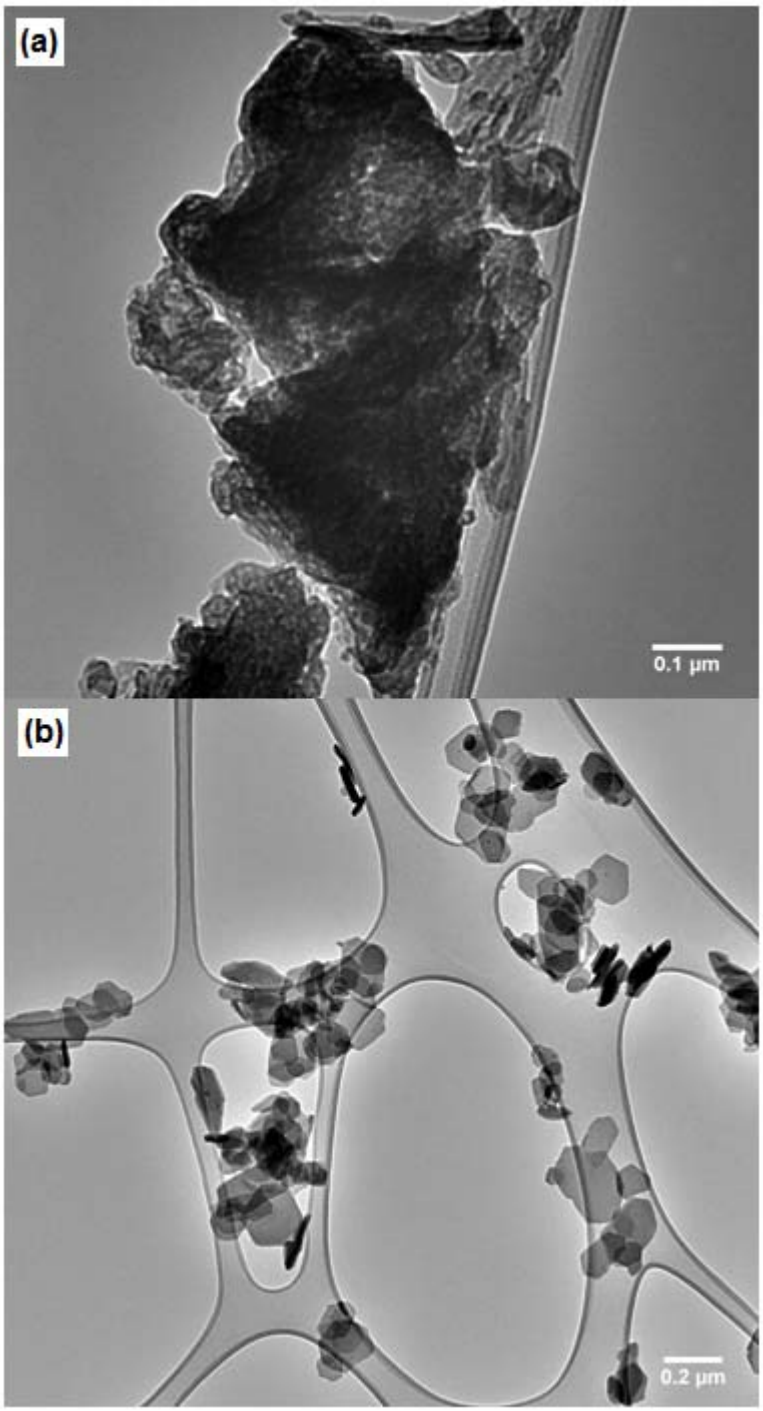

Figure S2. TEM image of acetone washed Zn_2Al-borate LDH after re-contacting with (a) H_2O and (b) Na_2CO_3 aqueous solution.

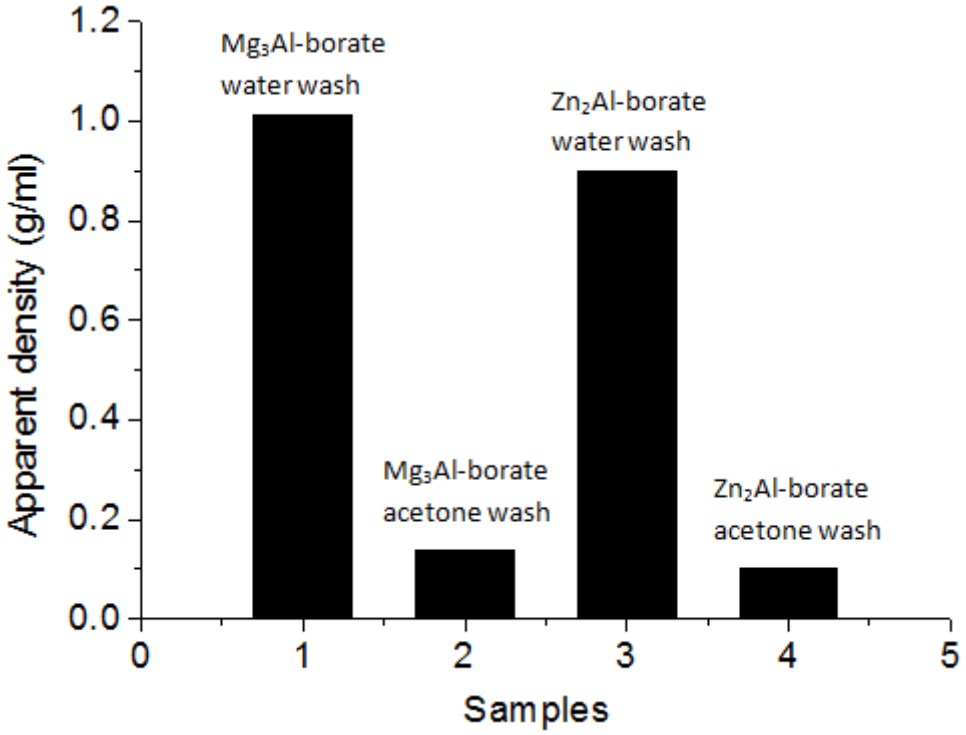

Figure S3. Apparent density of Mg_3Al-borate and Zn_2Al-borate washed with water or acetone.

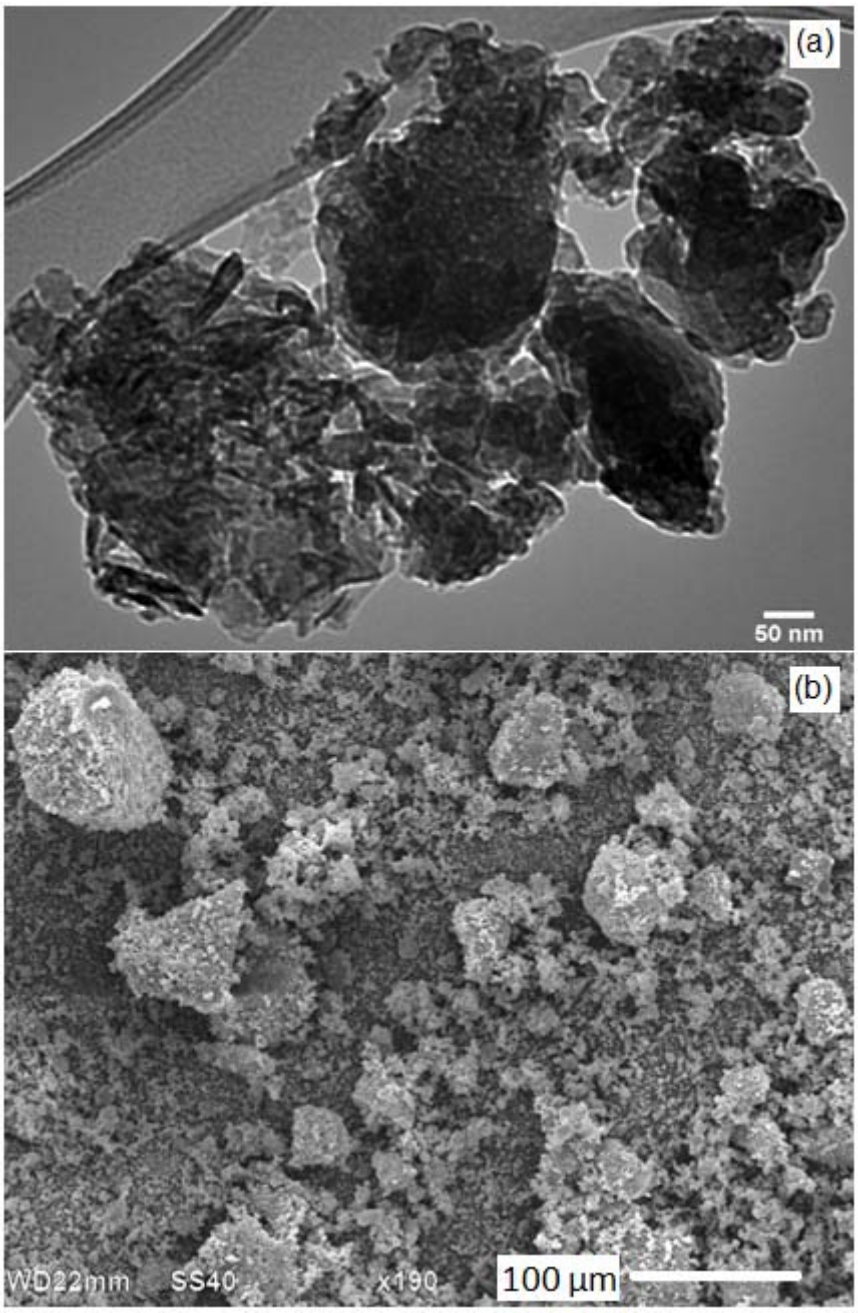

Figure S4. (a) TEM and (b) SEM images of Zn_2Al-borate LDH synthesized by conventional co-precipitation.

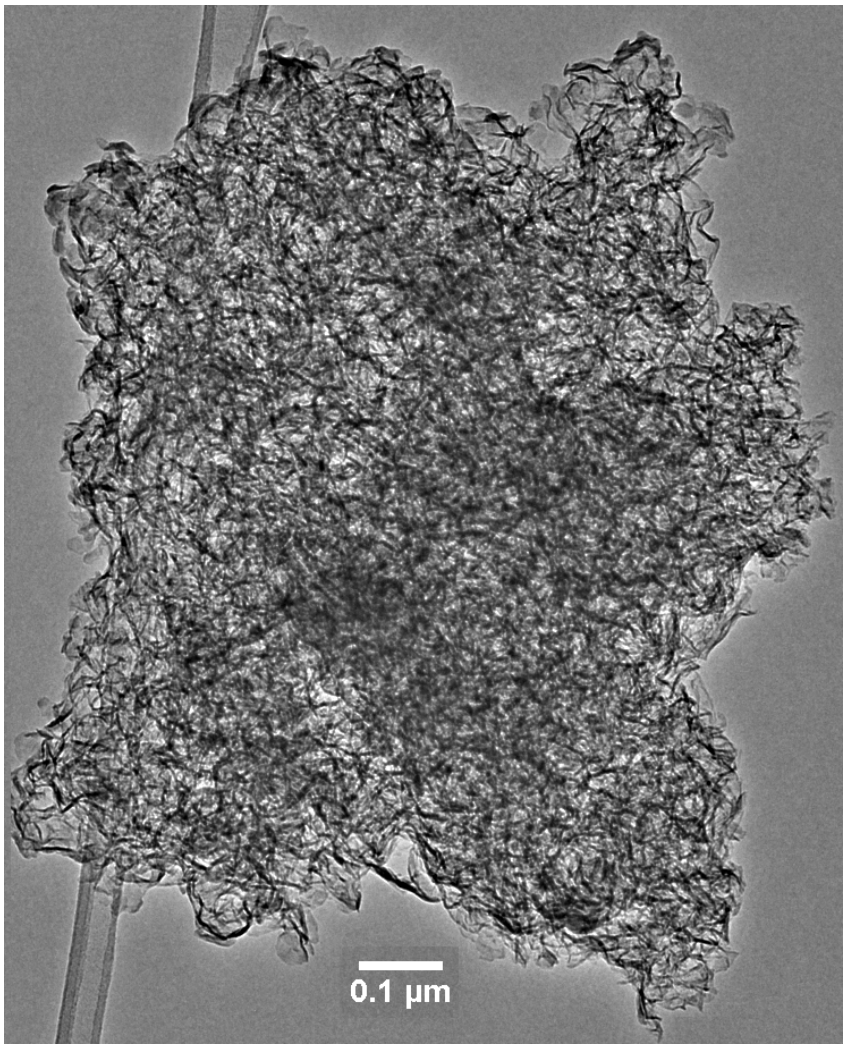

Figure S5. TEM image of Zn_2Al-borate LDH prepared by AMOST method.

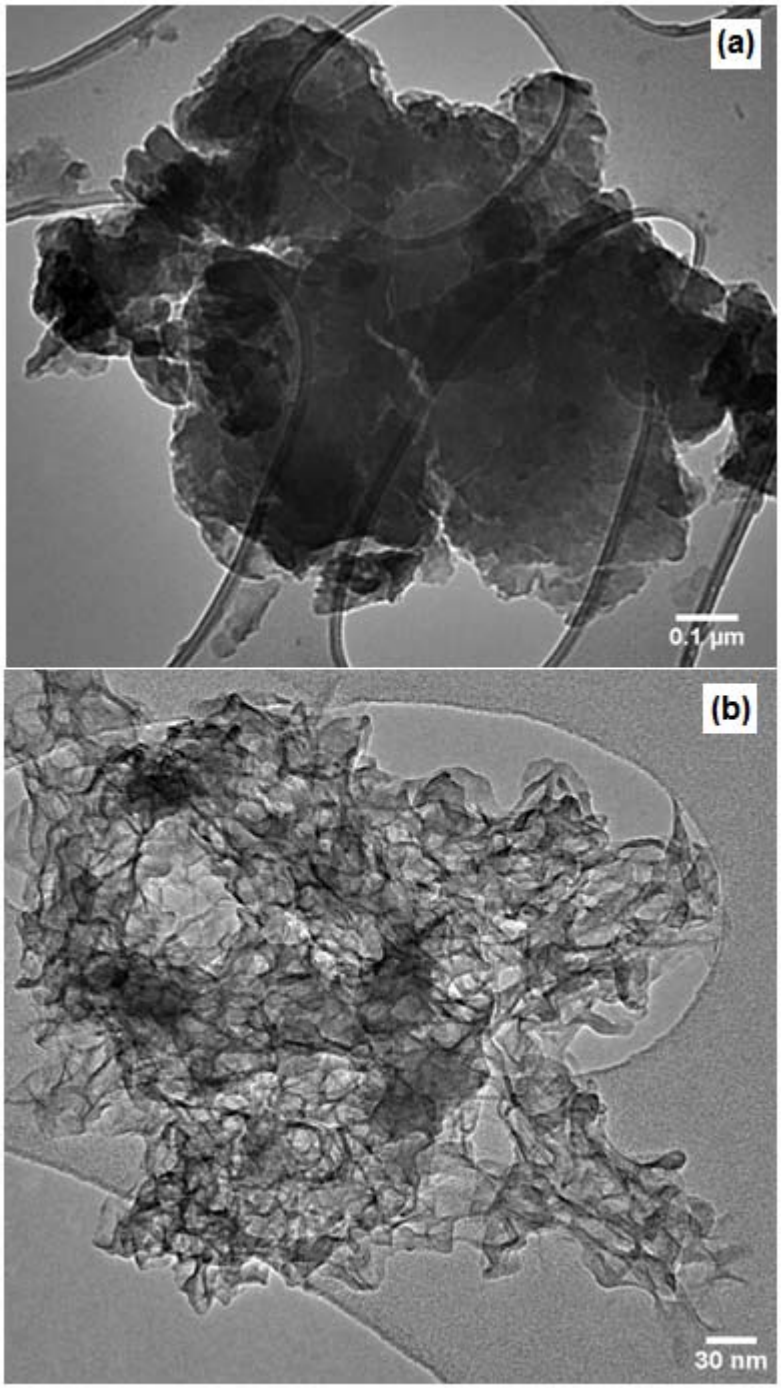

Figure S6. TEM images of Mg_3Al-borate LDHs washed with (a) water and (b) acetone.

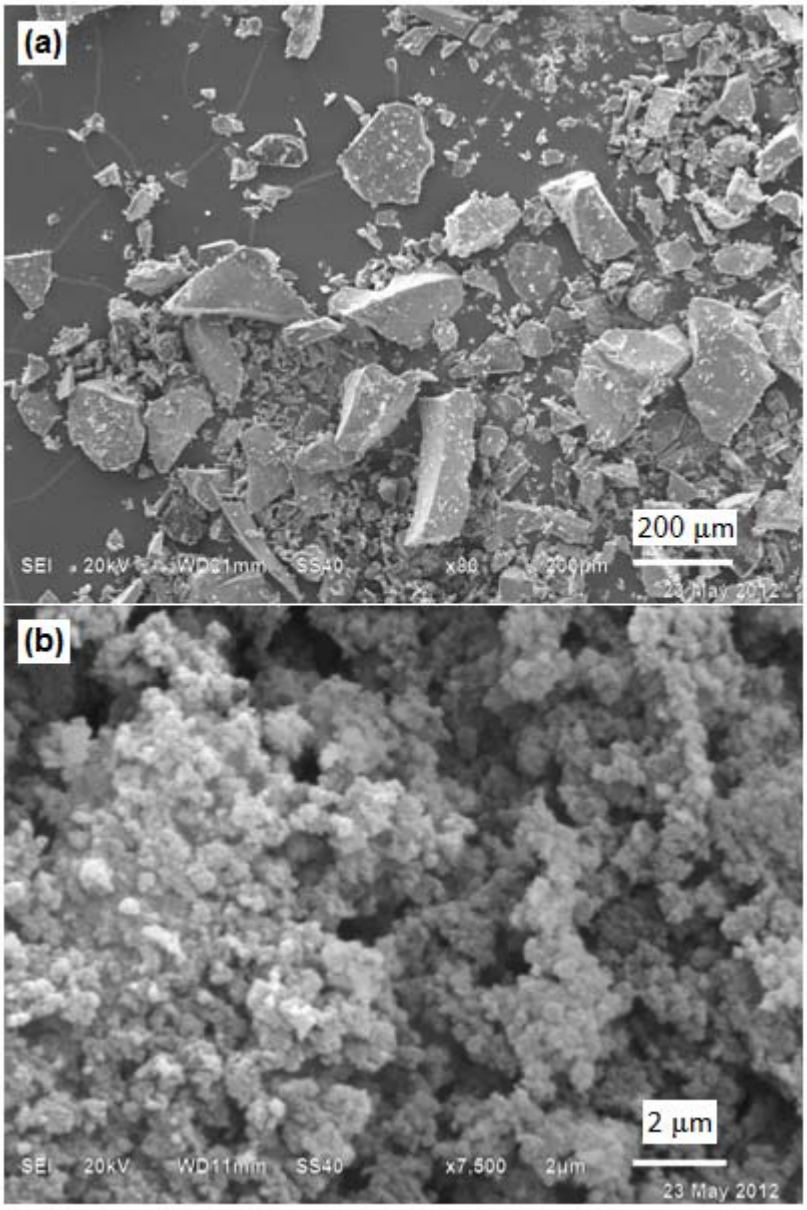

Figure S7. SEM images of Mg_3Al-borate LDH washed with (a) water and (b) acetone.

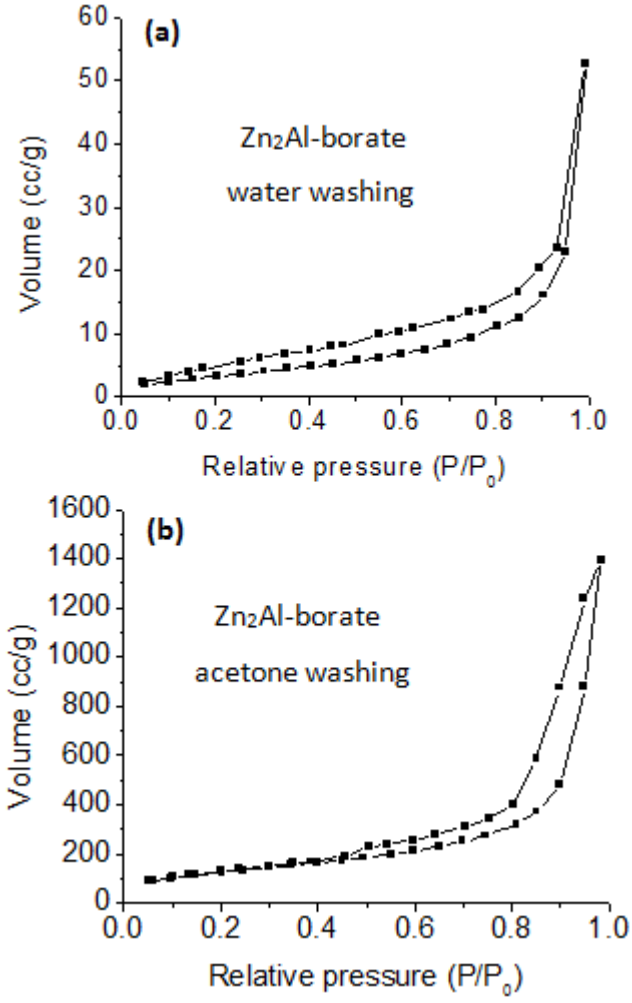

Figure S8. BET N_2 isotherms for Zn_2Al-borate LDHs washed with (a) water, and (b) acetone.

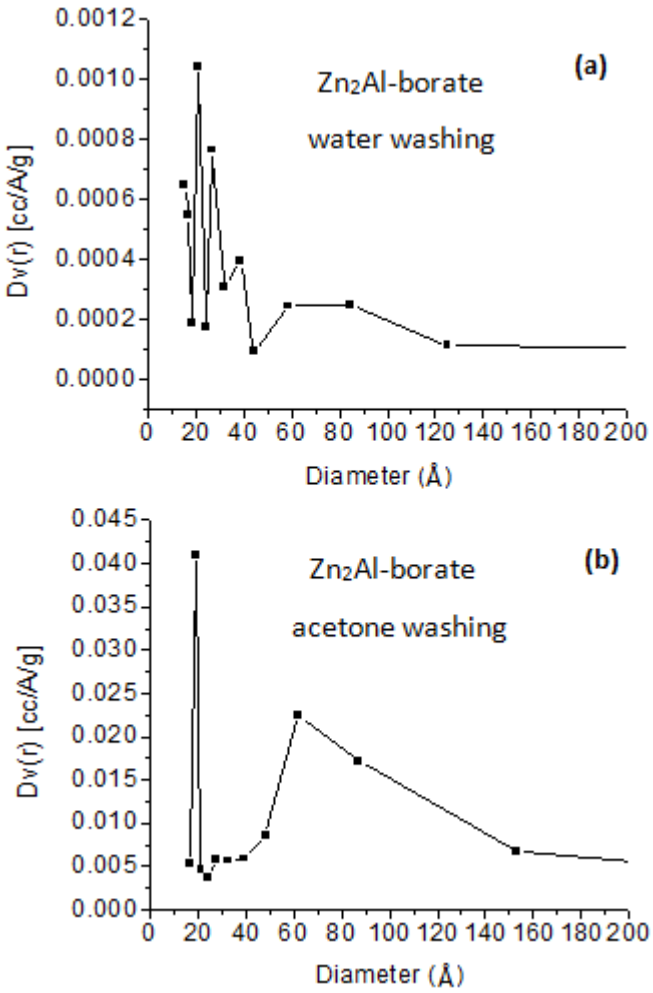

Figure S9. Pore size distribution of Zn_2Al-borate washed with (a) water and (b) acetone.

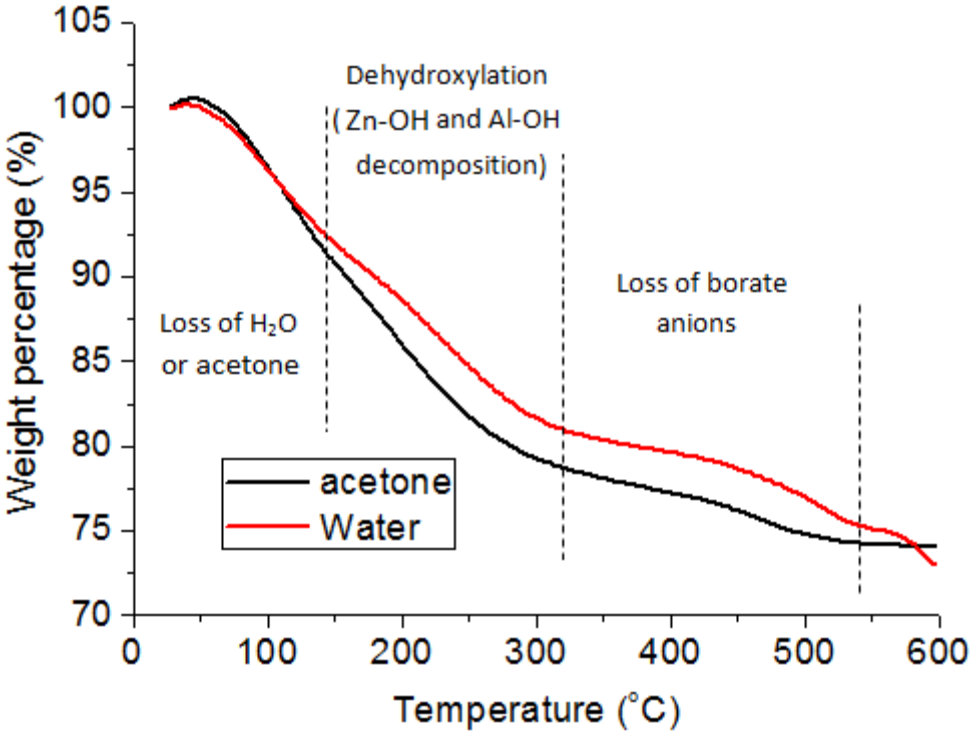

Figure S10. TGA analysis of Zn_2Al-borate washed with (a) acetone or (b) water.

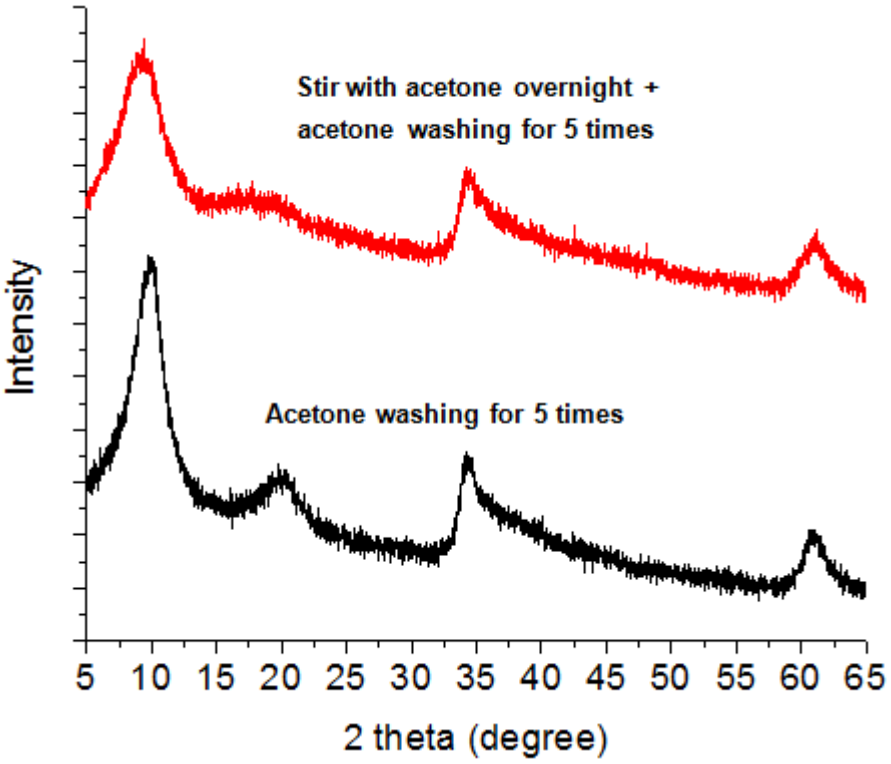

Figure S11. XRD patterns of water washed Zn_2Al-borate LDH dry powder that (a) stirred with acetone overnight + acetone washing for 5 times, and (b) acetone washing for 5 times.

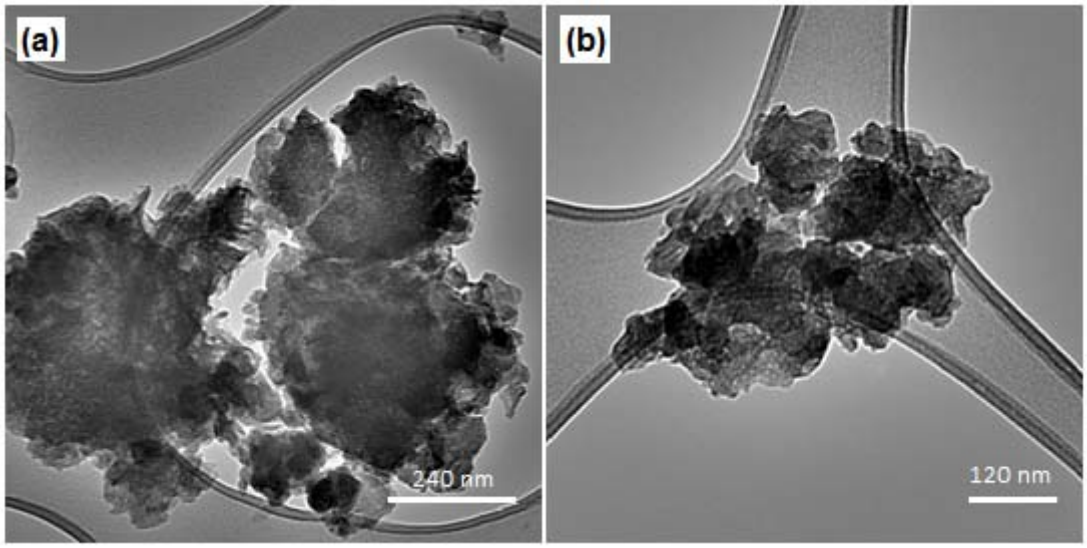

Figure S12. TEM images of water washed Zn_2Al-borate LDH dry powder that (a) stirred with acetone overnight + acetone washing for 5 times, and (b) acetone washing for 5 times.

Table S1. Summary of the M^{2+}/M^{3+} ratio, specific surface area, and total pore volume of synthesized Zn_2Al-borate and Mg_3Al-borate LDHs.

Samples	**M^{2+}/M^{3+}**	**Specific surface area (m^2/g)**	**Total pore volume (cc/g)**
Zn_2Al-borate (conventional)	1.13	13.4	0.08
Zn_2Al-borate (AMOST treatment)	1.04	458.6	2.15
Mg_3Al-borate (conventional)	1.77	1.0	0.01
Mg_3Al-borate (AMOST treatment)	1.86	263.0	1.07
Mg-Al-borate (reference 1)	1.20-1.60	60-80	×
Mg-Al-borate (reference 2)	×	20-40	×
Mg-Al-borate (reference 3)	×	47	×

1/3 References

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