

# 1 **Synthesis and Characterisation of Aqueous Miscible Organic-** 2 **Layered Double Hydroxides**

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## 1. Experimental details

### 1.1 The synthesis of Mg<sub>2</sub>Al-CO<sub>3</sub>-10 LDHs

The metal precursor solution (50 mL) of 0.75 M Mg(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O and 0.375 M Al(NO<sub>3</sub>)<sub>3</sub>·9H<sub>2</sub>O was added drop-wise into the 50 mL of 0.5 M Na<sub>2</sub>CO<sub>3</sub> base solution. The pH value was kept at *ca.* 10.0 by wise-dropping 4.0 M NaOH solution. This nucleation process usually takes 30 min. After aging for 16 h with stirring at room temperature, the mixture was filtered and washed with DI water until the pH was close to 7. Then the product was dried in the vacuum oven overnight. The final sample was named as Mg<sub>2</sub>Al-CO<sub>3</sub>-10-W. In the AMOST method, all the nucleation aging steps are the same as those in the conventional method. The LDH precursor was washed with DI water until the pH was close to 7 following by being rinsed with acetone thoroughly. The obtained LDH wet cake was dispersed in 200 mL acetone and stirred at room temperature for 1-2 h. Then the LDH was filtered and washed thoroughly with acetone again. Finally, the product was dried in the vacuum oven at room temperature for overnight. The sample was named as Mg<sub>2</sub>Al-CO<sub>3</sub>-10-A.

### 1.2 The synthesis of Mg<sub>3</sub>Al-CO<sub>3</sub>-12 LDHs

The metal precursor solution (50 mL) of 0.75 M Mg(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O and 0.25 M Al(NO<sub>3</sub>)<sub>3</sub>·9H<sub>2</sub>O was added drop-wise into the 50 mL of 0.5 M Na<sub>2</sub>CO<sub>3</sub> base solution. The pH value was kept at *ca.* 12.0 by wise-dropping 4.0 M NaOH solution. This nucleation process usually takes 30 min. After aging for 16 h with stirring at room temperature, the mixture was filtered and washed with DI water until the pH was close to 7. Then the product was dried in the vacuum oven overnight. The final sample was named as Mg<sub>3</sub>Al-CO<sub>3</sub>-12-W. In the AMOST method, all the nucleation aging steps are the same as those in the conventional method. The LDH precursor was washed with DI water until the pH was close to 7 following by being rinsed with acetone thoroughly. The obtained LDH wet cake was dispersed in 200 mL acetone and stirred at room temperature for 1-2 h. Then the LDH was filtered and washed thoroughly with acetone again. Finally, the product was dried in the vacuum oven at room temperature for overnight. The sample was named as Mg<sub>3</sub>Al-CO<sub>3</sub>-12-A. When method was used as AMO solvent, the final sample name is Mg<sub>3</sub>Al-CO<sub>3</sub>-12-M.

### 1.3 The synthesis of Mg<sub>3</sub>Al-NO<sub>3</sub>-10 LDHs

The metal precursor solution (50 mL) of 0.75 M Mg(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O and 0.25 M Al(NO<sub>3</sub>)<sub>3</sub>·9H<sub>2</sub>O was added drop-wise into the 50 mL of 0.5 M NaNO<sub>3</sub> base solution. The pH value was kept at *ca.* 10.0 by wise-dropping 4.0 M NaOH solution. During this synthesis, the system was purged with N<sub>2</sub> gas to prevent the contamination by atmospheric CO<sub>2</sub>. This nucleation process usually takes 30 min. After aging for 16 h with stirring at room temperature, the mixture was filtered and washed with DI water until the pH was close to 7. Then the product was dried in the vacuum oven overnight. The final sample was named as Mg<sub>3</sub>Al-NO<sub>3</sub>-10-W. In the AMOST method, all the nucleation aging steps are the same as those in the conventional method. The LDH precursor was washed with DI water until the pH was close to 7 following by being rinsed with acetone thoroughly. The obtained LDH wet cake was dispersed in 200 mL acetone and stirred at room temperature for 1 - 2 h. Then the

1 LDH was filtered and washed thoroughly with acetone again. Finally, the product was dried  
2 in the vacuum oven at room temperature for overnight. The sample was named as Mg<sub>3</sub>Al-  
3 NO<sub>3</sub>-10-A.

#### 4 1.4 The synthesis of Mg<sub>3</sub>Al-SO<sub>4</sub>-10 LDHs

5 The metal precursor solution (50 mL) of 0.75 M MgSO<sub>4</sub>·7H<sub>2</sub>O and 0.25 M Al<sub>3</sub>(SO<sub>4</sub>)<sub>2</sub>·16H<sub>2</sub>O  
6 was added drop-wise into the 50 mL of 0.5 M Na<sub>2</sub>SO<sub>4</sub> base solution. The pH value was kept  
7 at *ca.* 10.0 by wise-dropping 4.0 M NaOH solution. During the synthesis, the system was  
8 purged with N<sub>2</sub> gas to prevent the contamination by atmospheric CO<sub>2</sub>. This nucleation  
9 process usually takes 30 min. After aging for 16 h with stirring at room temperature, the  
10 mixture was filtered and washed with DI water until the pH was close to 7. Then the product  
11 was dried in the vacuum oven overnight. The final sample was named as Mg<sub>3</sub>Al-SO<sub>4</sub>-10-W.  
12 In the AMOST method, all the nucleation aging steps are the same as those in the  
13 conventional method. The LDH precursor was washed with DI water until the pH was close  
14 to 7 following by being rinsed with acetone thoroughly. The obtained LDH wet cake was  
15 dispersed in 200 mL acetone and stirred at room temperature for 1 - 2 h. Then the LDH was  
16 filtered and washed thoroughly with acetone again. Finally, the product was dried in the  
17 vacuum oven at room temperature for overnight. The sample was named as Mg<sub>3</sub>Al-SO<sub>4</sub>-10-A.

#### 18 1.5 The synthesis of Mg<sub>3</sub>Al-Cl-10 LDHs

19 The metal precursor solution (50 mL) of 0.75 M MgCl<sub>2</sub>·6H<sub>2</sub>O and 0.25 M AlCl<sub>3</sub>·6H<sub>2</sub>O was  
20 added drop-wise into the 50 mL of 0.5 M NaCl base solution. The pH value was kept at *ca.*  
21 10.0 by wise-dropping 4.0 M NaOH solution. During the synthesis, the system was purged  
22 with N<sub>2</sub> gas to prevent the contamination by atmospheric CO<sub>2</sub>. This nucleation process  
23 usually takes 30 min. After aging for 16 h with stirring at room temperature, the mixture was  
24 filtered and washed with DI water until the pH was close to 7. Then the product was dried in  
25 the vacuum oven overnight. The final sample was named as Mg<sub>3</sub>Al-Cl-10-W. In the AMOST  
26 method, all the nucleation aging steps are the same as those in the conventional method. The  
27 LDH precursor was washed with DI water until the pH was close to 7 following by being  
28 rinsed with acetone thoroughly. The obtained LDH wet cake was dispersed in 200 mL  
29 acetone and stirred at room temperature for 1 - 2 h. Then the LDH was filtered and washed  
30 thoroughly with acetone again. Finally, the product was dried in the vacuum oven at room  
31 temperature for overnight. The sample was named as Mg<sub>3</sub>Al-Cl-10-A.

#### 32 1.6 The synthesis of Mg<sub>3</sub>Al<sub>0.5</sub>Fe<sub>0.5</sub>-NO<sub>3</sub> -10 LDHs

33 The metal precursor solution (50 mL) of 0.75 M Mg(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O, 0.125 M Al(NO<sub>3</sub>)<sub>3</sub>·9H<sub>2</sub>O  
34 and 0.125 M Fe(NO<sub>3</sub>)<sub>3</sub>·9H<sub>2</sub>O was added drop-wise into the 50 mL of 0.5 M NaNO<sub>3</sub> base  
35 solution. The pH value was kept at *ca.* 10.0 by wise-dropping 4.0 M NaOH solution. During  
36 the synthesis, the system was purged with N<sub>2</sub> gas to prevent the contamination by  
37 atmospheric CO<sub>2</sub>. This nucleation process usually takes 30 min. After aging for 16 h with  
38 stirring at room temperature, the mixture was filtered and washed with DI water until the pH  
39 was close to 7. Then the product was dried in the vacuum oven overnight. The final sample  
40 was named as Mg<sub>3</sub>Al<sub>0.5</sub>Fe<sub>0.5</sub>-NO<sub>3</sub> -10-W. In the AMOST method, all the nucleation aging

1 steps are the same as those in the conventional method. The LDH precursor was washed with  
2 DI water until the pH was close to 7 following by being rinsed with acetone thoroughly. The  
3 obtained LDH wet cake was dispersed in 200 mL acetone and stirred at room temperature for  
4 1 - 2 h. Then the LDH was filtered and washed thoroughly with acetone again. Finally, the  
5 product was dried in the vacuum oven at room temperature for overnight. The sample was  
6 named as  $\text{Mg}_3\text{Al}_{0.5}\text{Fe}_{0.5}\text{-NO}_3$  -10-A.

## 7 1.7 The synthesis of $\text{Zn}_2\text{Al}$ -Borate-8.3 and $\text{Mg}_3\text{Al}$ -Borate-9 LDHs

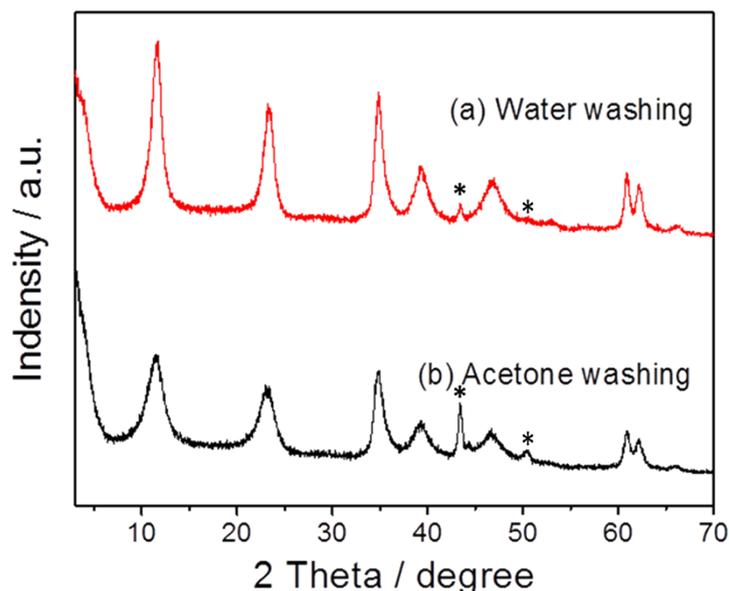
8 The synthesis of  $\text{Zn}_2\text{Al}$ -Borate-8.3 and  $\text{Mg}_3\text{Al}$ -Borate-9 was according to our previous  
9 report<sup>[1]</sup>. The metal precursor solution (50 mL) of 0.75 M  $\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  and 0.375 M  
10  $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$  was added drop-wise into the 50 mL of 1.87 M  $\text{H}_3\text{BO}_3$  solution base solution.  
11 The pH value was kept at *ca.* 8.3 by wise-dropping 1.0 M NaOH solution. During the  
12 synthesis, the system was purged with  $\text{N}_2$  gas to prevent the contamination by atmospheric  
13  $\text{CO}_2$ . This nucleation process usually takes 30 min. After aging for 16 h with stirring at 65 °C,  
14 the mixture was filtered and washed with DI water until the pH was close to 7. Then the  
15 product was dried in the vacuum oven overnight. The final sample was named as  $\text{Zn}_2\text{Al}$ -  
16 Borate-8.3-10-W. In the AMOST method, all the nucleation aging steps are the same as those  
17 in the conventional method. The LDH precursor was washed with DI water until the pH was  
18 close to 7 following by being rinsed with acetone thoroughly. The obtained LDH wet cake  
19 was dispersed in 200 mL acetone and stirred at room temperature for 1 - 2 h. Then the LDH  
20 was filtered and washed thoroughly with acetone again. Finally, the product was dried in the  
21 vacuum oven at room temperature for overnight. The sample was named as  $\text{Zn}_2\text{Al}$ -Borate-  
22 8.3-A.

23 Similarly,  $\text{Mg}_3\text{Al}$ -borate LDH was synthesized at pH 9 using 50 mL of 0.75 M  
24  $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  and 0.25 M  $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$  solution drop-wise into a 50 mL of 1.87 M  
25  $\text{H}_3\text{BO}_3$  solution. During the synthesis, the system was purged with  $\text{N}_2$  gas to prevent the  
26 contamination by atmospheric  $\text{CO}_2$ . This nucleation process usually takes us 30 min. After  
27 aging for 16 h with stirring at 65 °C, the mixture was filtered and washed with DI water until  
28 the pH was close to 7. Then the mixture was dried in the vacuum oven overnight. The final  
29 sample was named as  $\text{Mg}_3\text{Al}$ -Borate-9-W. All the previous steps are the same with the  
30 conventional method except the water washed LDHs wet cake was re-dispersed in 200 mL  
31 acetone and stirred at room temperature for 1 - 2 h several times. Then the LDH was filtered  
32 and washed thoroughly with acetone again. Finally, the product was dried in the vacuum  
33 oven at room temperature for overnight. The sample was named  $\text{Mg}_3\text{Al}$ -Borate-9-A.

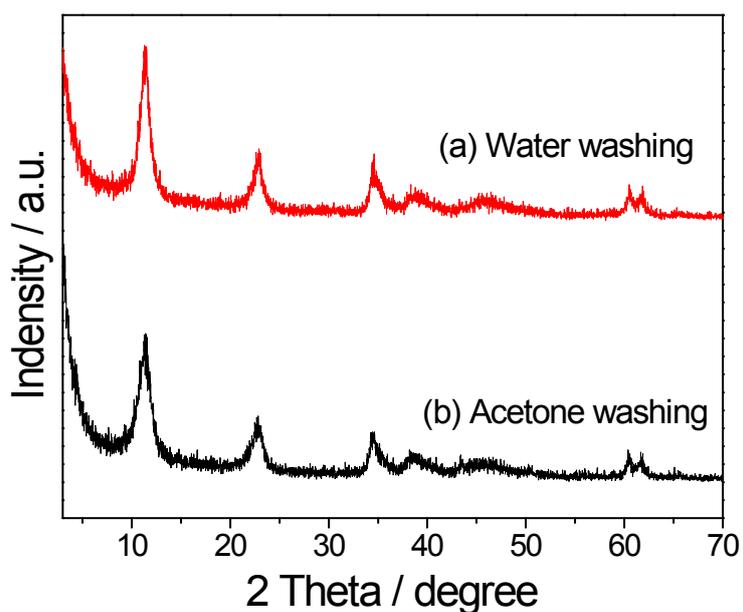
## 34 2. Characterization

### 35 2.1 X-ray diffraction

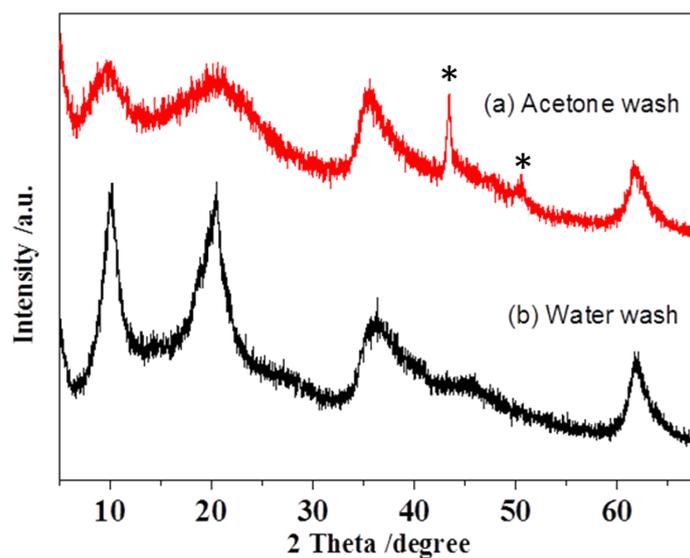
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1  
 2 **Fig. S1** XRD patterns of  $Mg_2Al-CO_3-10$  (a) sample prepared by conventional co-precipitation  
 3 method in water at pH 10 (b) sample prepared under identical synthesis conditions with the  
 4 additional AMOST method treatment using acetone as the AMO-solvent. (\*) are Bragg  
 5 reflections from the Al sample holder.  
 6

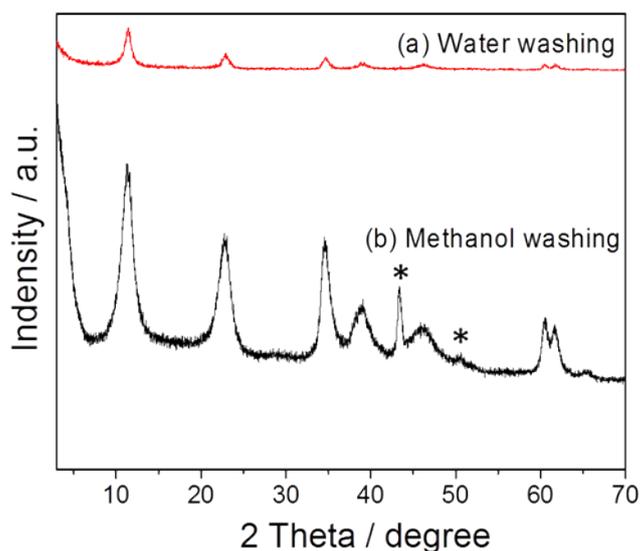


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 8 **Fig. S2** XRD patterns of  $Mg_3Al-CO_3-12$  (a) sample prepared by conventional co-precipitation  
 9 method in water at pH 12 (b) sample prepared under identical synthesis conditions with the  
 10 additional AMOST method treatment using acetone as the AMO-solvent.  
 11



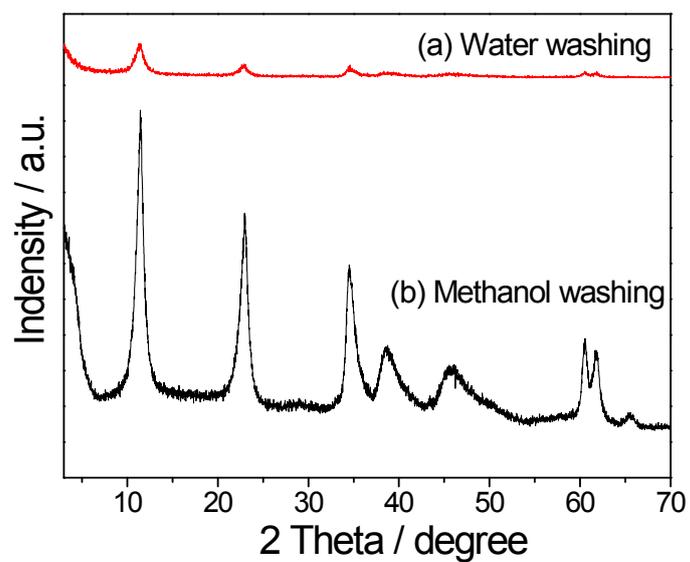
1  
 2 **Fig. S3** XRD patterns of  $\text{Mg}_3\text{Al-SO}_4\text{-10}$  (a) sample prepared by conventional co-precipitation  
 3 method in water at pH 10 (b) sample prepared under identical synthesis conditions with the  
 4 additional AMOST method treatment using acetone as the AMO-solvent. (\*) are Bragg  
 5 reflections from the Al sample holder.

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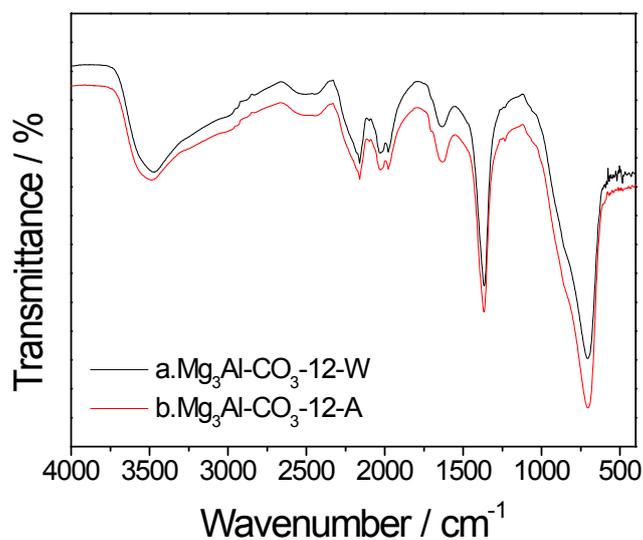
9  
 10 **Fig. S4** XRD patterns of  $\text{Mg}_3\text{Al-CO}_3\text{-10}$  (a) sample prepared by conventional co-precipitation  
 11 method in water at pH 10 (b) sample prepared under identical synthesis conditions with the  
 12 additional AMOST method treatment using methanol as the AMO-solvent. (\*) are Bragg  
 13 reflections from the Al sample holder

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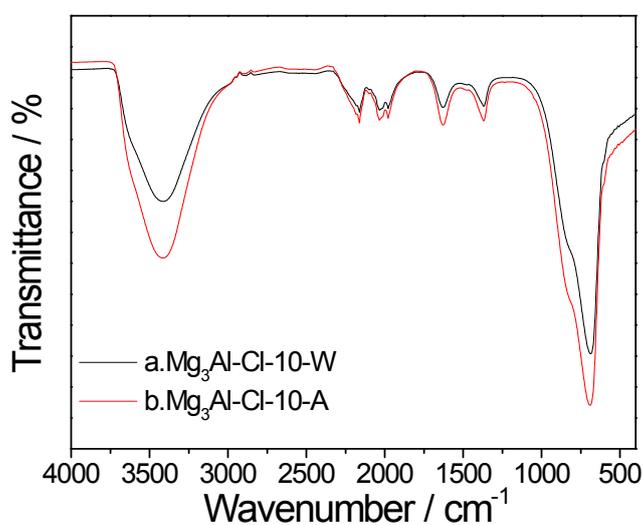


1  
2 **Fig. S5** XRD patterns of Mg<sub>3</sub>Al-SO<sub>4</sub>-10 (a) sample prepared by conventional co-precipitation  
3 method in water at pH 10 (b) sample prepared under identical synthesis conditions with the  
4 additional AMOST method treatment using methanol as the AMO-solvent.  
5

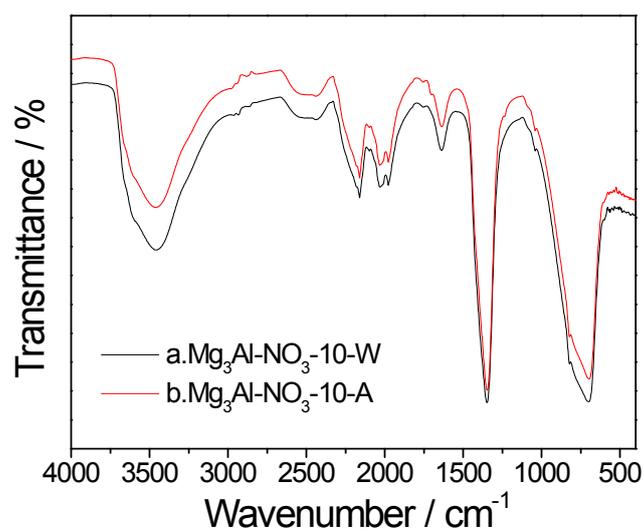
## 1 2.2 Infrared spectroscopy



2  
3 **Fig. S6** FTIR patterns of Mg<sub>3</sub>Al-CO<sub>3</sub>-12 (a) sample prepared by conventional co-  
4 precipitation method in water at pH 10 (b) sample prepared under identical synthesis  
5 conditions with the additional AMOST method treatment using acetone as the AMO-solvent.  
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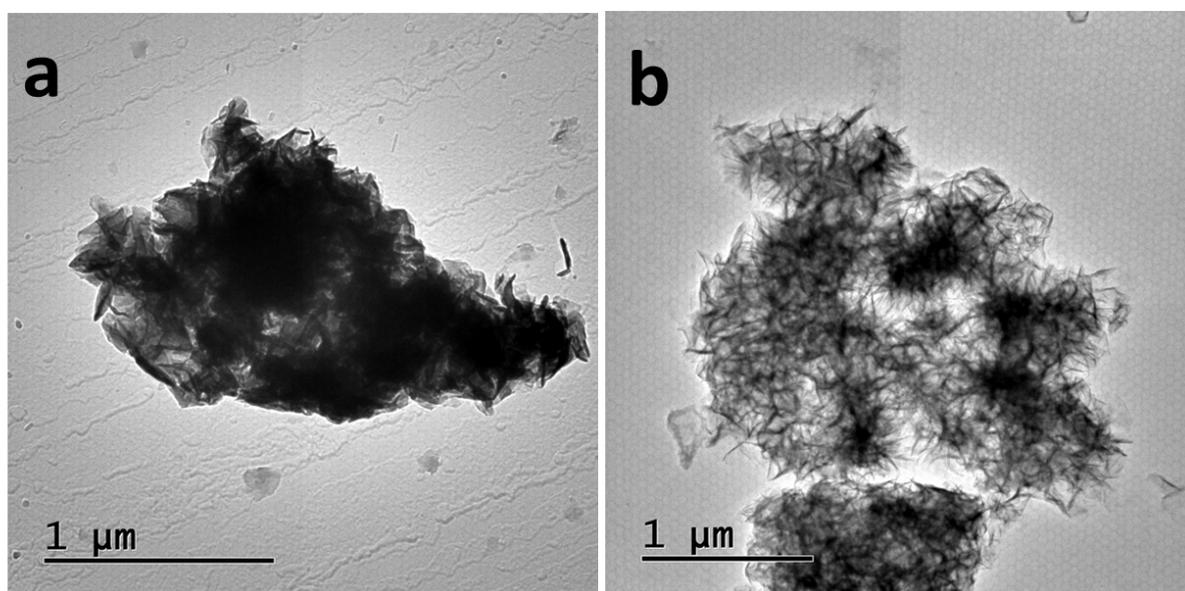
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8 **Fig. S7** FTIR patterns of Mg<sub>3</sub>Al-Cl-10 (a) sample prepared by conventional co-precipitation  
9 method in water at pH 10 (b) sample prepared under identical synthesis conditions with the  
10 additional AMOST method treatment using acetone as the AMO-solvent.



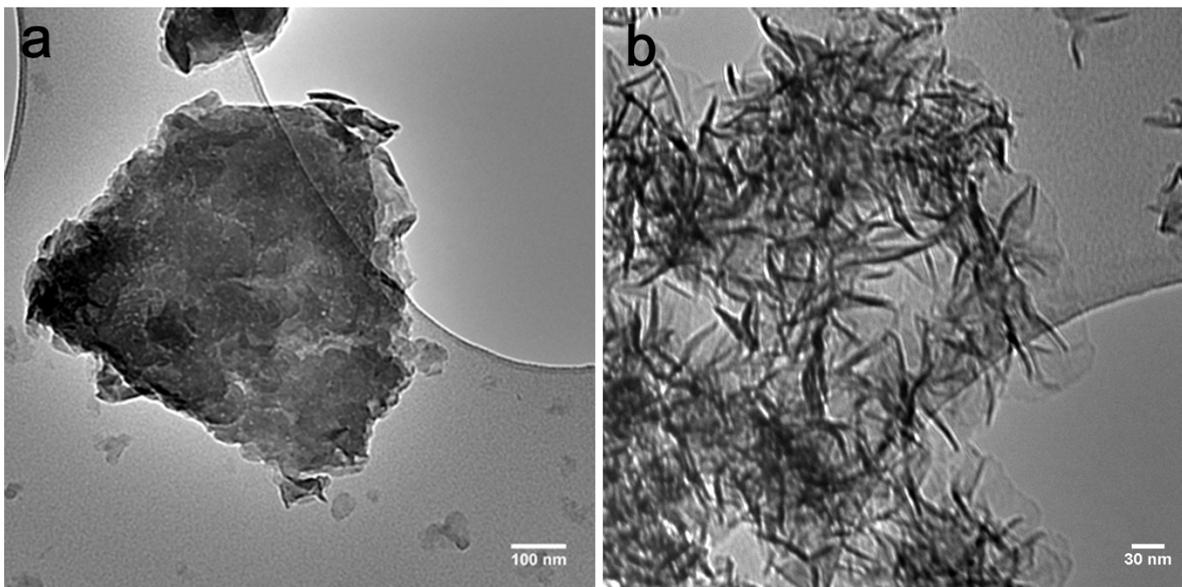
1  
 2 **Fig. S8** FTIR patterns of  $\text{Mg}_3\text{Al-NO}_3\text{-10}$  (a) sample prepared by conventional co-  
 3 precipitation method in water at pH 10 (b) sample prepared under identical synthesis  
 4 conditions with the additional AMOST method treatment using acetone as the AMO-solvent.  
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### 6 **2.3** Transmission Electron Microscopy

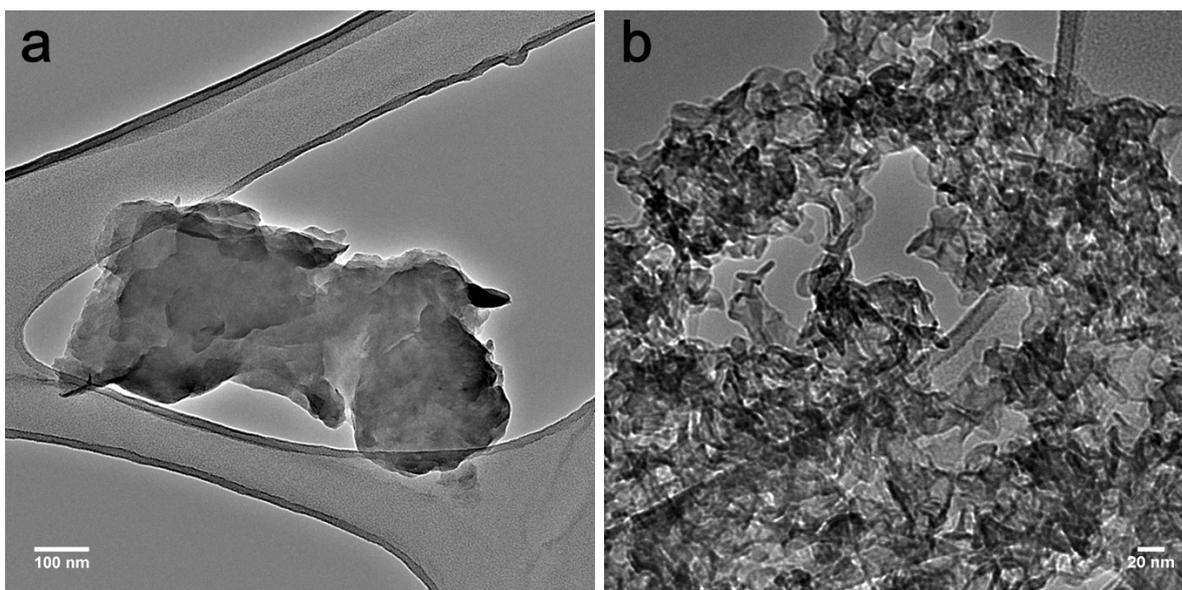
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 9 **Fig. S9** TEM patterns of  $\text{Mg}_3\text{Al-Cl-10}$  (a) sample prepared by conventional co-precipitation  
 10 method in water at pH 10 (b) sample prepared under identical synthesis conditions with the  
 11 additional AMOST method treatment using acetone as the AMO-solvent.  
 12

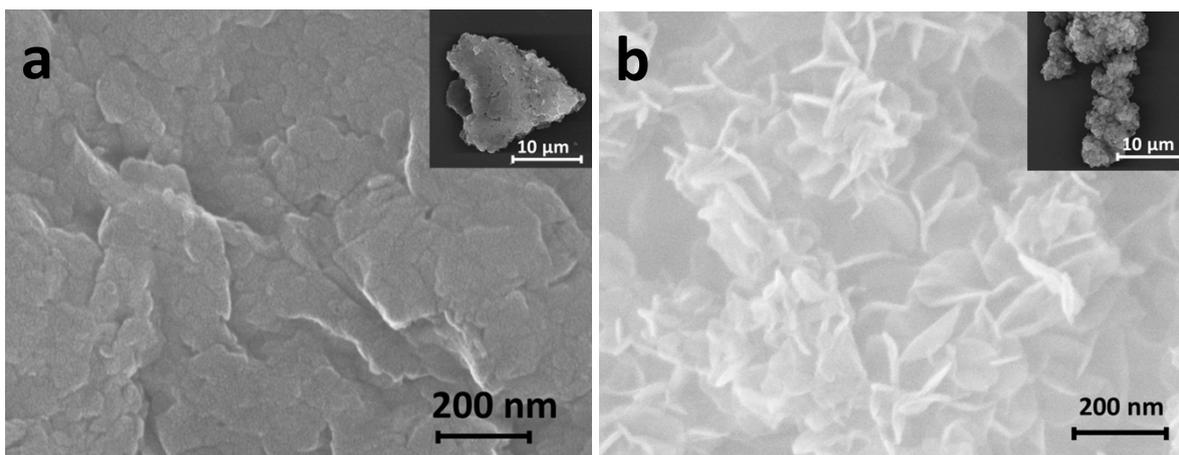


1  
2 **Fig. S10** TEM patterns of  $Mg_3Al-NO_3-10$  (a) sample prepared by conventional co-  
3 precipitation method in water at pH 10 (b) sample prepared under identical synthesis  
4 conditions with the additional AMOST method treatment using acetone as the AMO-solvent.  
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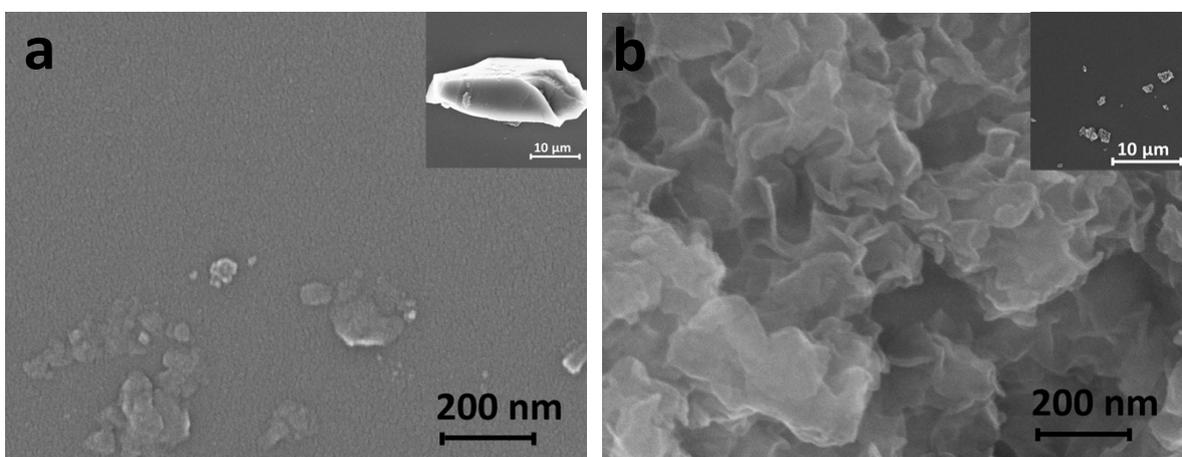
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8 **Fig. S11** TEM patterns of  $Mg_3Al-SO_4-10$  (a) sample prepared by conventional co-  
9 precipitation method in water at pH 10 (b) sample prepared under identical synthesis  
10 conditions with the additional AMOST method treatment using acetone as the AMO-solvent.  
11

12  
13 **2.4 Scanning Electron Microscopy**



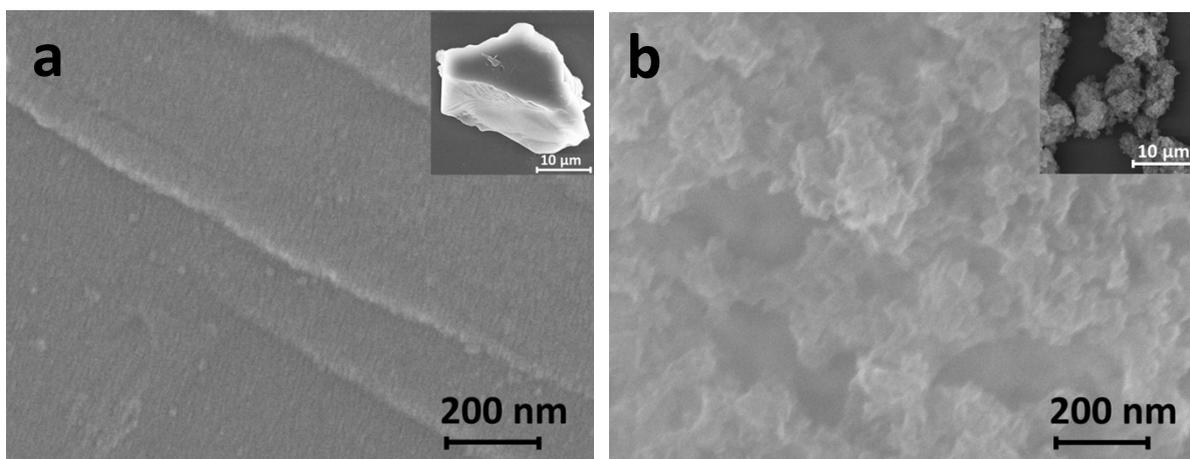
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3 **Fig. S12** TEM patterns of  $\text{Mg}_3\text{Al-Cl-10}$  (a) sample prepared by conventional co-precipitation  
4 method in water at pH 10 (b) sample prepared under identical synthesis conditions with the  
5 additional AMOST method treatment using acetone as the AMO-solvent.



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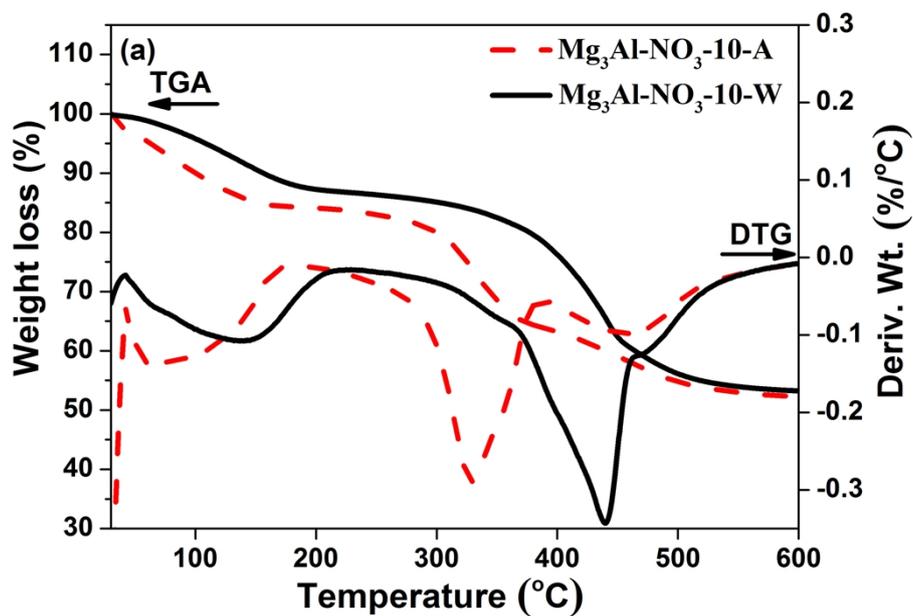
8 **Fig. S13** TEM patterns of  $\text{Mg}_3\text{Al-NO}_3\text{-10}$  (a) sample prepared by conventional co-  
9 precipitation method in water at pH 10 (b) sample prepared under identical synthesis  
10 conditions with the additional AMOST method treatment using acetone as the AMO-solvent.



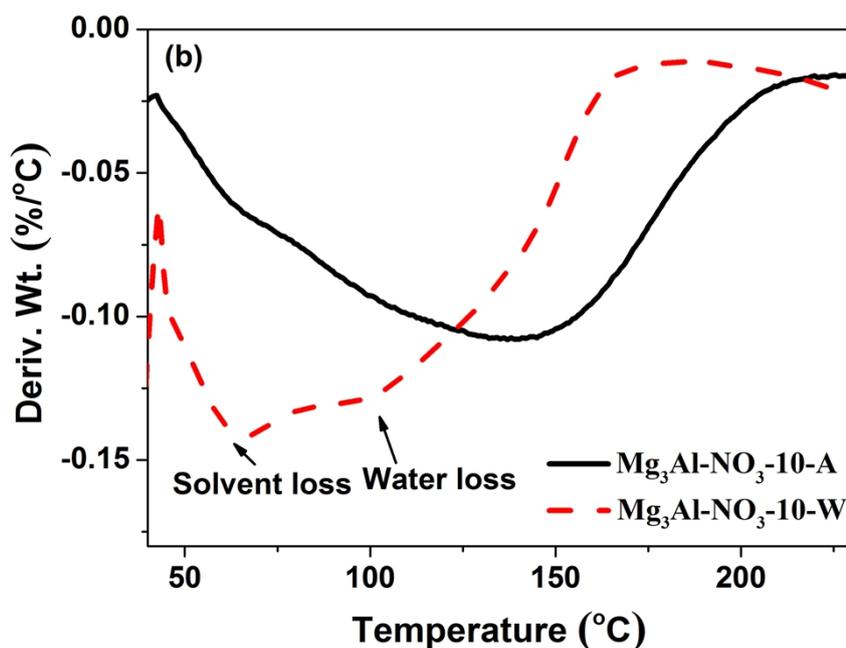
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**Fig. S14** TEM patterns of  $\text{Mg}_3\text{Al-SO}_4\text{-10}$  (a) sample prepared by conventional co-  
precipitation method in water at pH 10 (b) sample prepared under identical synthesis  
conditions with the additional AMOST method treatment using acetone as the AMO-solvent.

## 1 2.5 Thermogravimetric analysis



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4 **Fig. S15.** TGA and DTG analysis of Mg<sub>3</sub>Al-NO<sub>3</sub>-10 LDHs (a) in the range of 30-600 °C; (b)  
5 in the range of 30-230 °C; Mg<sub>3</sub>Al-NO<sub>3</sub>-10-W prepared by a conventional co-precipitation  
6 method in water at pH 10. Mg<sub>3</sub>Al-NO<sub>3</sub>-10-A is prepared by identical conditions in water at  
7 pH 10 according to the AMOST method using acetone as the AMO-solvent.

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2 **Table S1** Summary of water and AMO-solvent content in the AMO-LDHs compared to  
 3 conventional C-LDHs as determined by analysis of the TGA data.

LDH	C-LDH <sup>2</sup>		AMO-LDH-A <sup>1</sup>		AMO-LDH-M <sup>1</sup>	
	<i>b</i> <sup>3</sup>	<i>c</i> <sup>4</sup>	<i>b</i> <sup>3</sup>	<i>c</i> <sup>4</sup>	<i>b</i> <sup>3</sup>	<i>c</i> <sup>4</sup>
<b>Mg<sub>3</sub>Al-CO<sub>3</sub>-10</b>	0.41	0	0.34	0.04	--	--
<b>Mg<sub>3</sub>Al-CO<sub>3</sub>-12</b>	0.7	0	0.43	0.11	0.44	0.11
<b>Mg<sub>2</sub>Al-CO<sub>3</sub>-10</b>	1.05	0	0.59	0.18	--	--
<b>Mg<sub>3</sub>Al-Cl-10</b>	0.58	0	0.46	0.04	--	--
<b>Mg<sub>3</sub>Al-SO<sub>4</sub>-10</b>	0.77	0	0.71	0.17	--	--
<b>Mg<sub>3</sub>Al-NO<sub>3</sub>-10</b>	0.57	0	0.38	0.12	--	--
<b>Mg<sub>3</sub>Al<sub>0.5</sub>Fe<sub>0.5</sub>-10</b>	0.74	0	0.5	0.06	--	--

4 <sup>1</sup>AMO-LDH-A and AMO-LDH-M are the LDH with the formula of  $[M^{z+}_{1-x}M'^{y+}_x(OH)_2]^{a+}(X^{n-})_{a/r} \cdot bH_2O \cdot c(AMO-solvent)$  (**1**); wherein M and M' are metal cations,  $z = 1$  or  $2$ ;  $y = 3$  or  $4$ ,  $0 < x < 1$ ,  $b = 0-10$ ,  $c = 0-10$ , X is an anion,  $r = 1$  to  $3$  and  $a = z(1-x) + xy - 2$ . AMO-solvent (A =  
 5 Acetone, M = Methanol).  
 6  
 7

8 <sup>2</sup>C-LDH is an LDH with the formula  $[M^{z+}_{1-x}M'^{y+}_x(OH)_2]^{a+}(X^{n-})_{a/r} \cdot bH_2O$  (**2**); wherein M and  
 9 M' are metal cations,  $z = 1$  or  $2$ ;  $y = 3$  or  $4$ ,  $0 < x < 1$ ,  $b = 0-10$ ,  $c = 0-10$ , X is an anion,  $r = 1$  to  $3$   
 10 and  $a = z(1-x) + xy - 2$ .

11 <sup>3</sup>*b* is the water content in the formula (**1**) and (**2**).

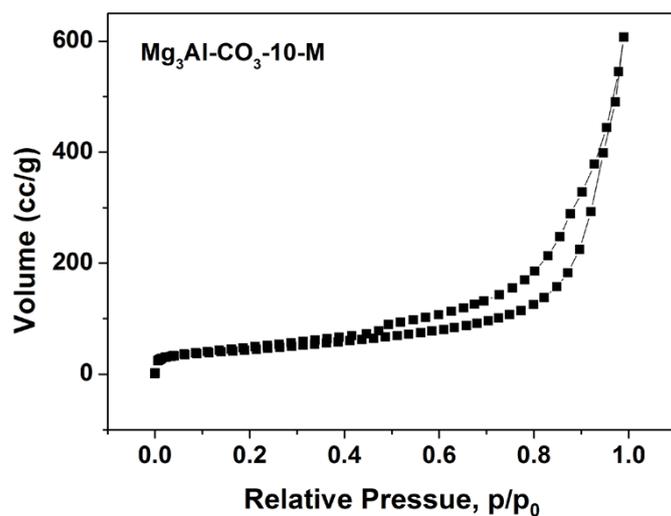
12 <sup>4</sup>*c* is the acetone content in the formula (**1**).

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## 1 2.6 BET Analysis

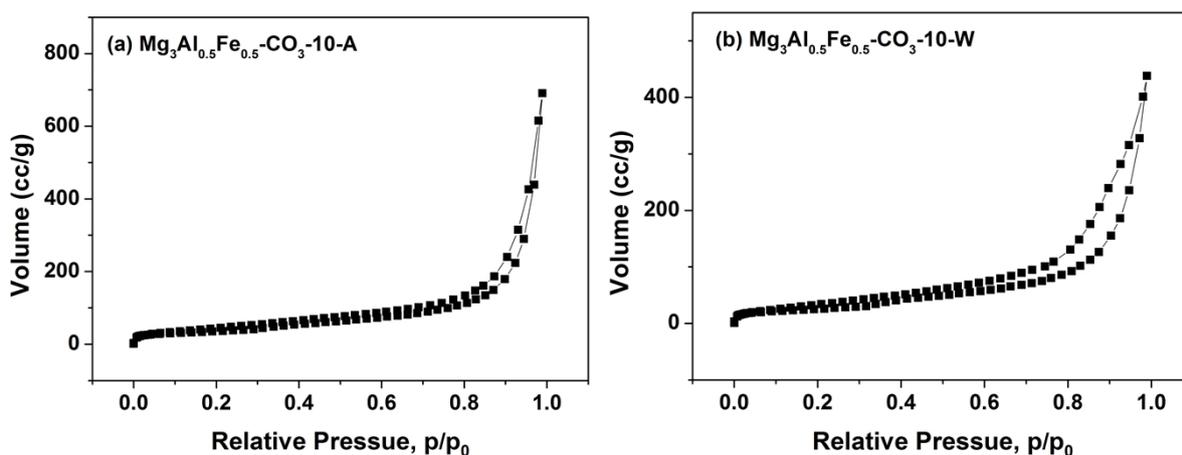


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4 **Fig. S16** BET Isotherms of  $\text{Mg}_3\text{Al-CO}_3\text{-10}$  BET sample prepared under identical synthesis  
5 conditions with the additional AMOST method treatment using methanol as the AMO-  
6 solvent.

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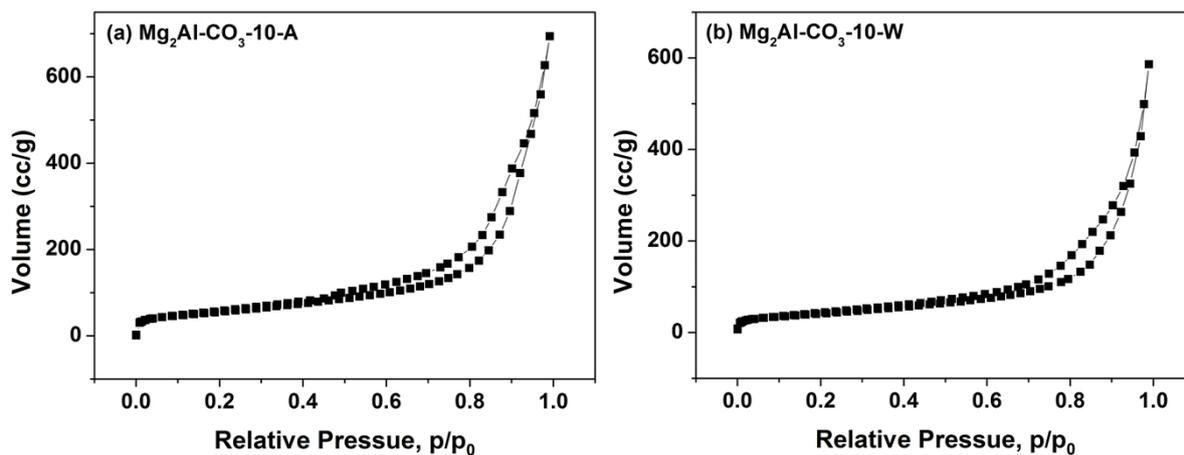
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10 **Fig. S17** BET Isotherms of  $\text{Mg}_3\text{Al}_{0.5}\text{Fe}_{0.5}\text{-CO}_3\text{-10}$  (a) sample prepared under identical  
11 synthesis conditions with the additional AMOST method treatment using acetone as the  
12 AMO-solvent 10 (b) sample prepared by conventional co-precipitation method in water at  
13 pH.

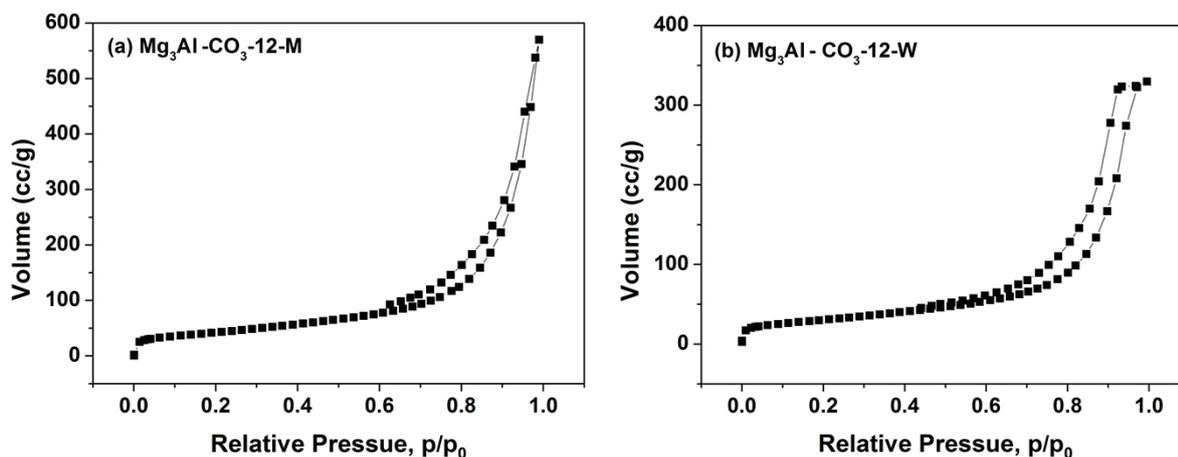
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 2 **Fig. S18** BET Isotherms of  $\text{Mg}_2\text{Al-CO}_3\text{-10}$  (a) sample prepared under identical synthesis  
 3 conditions with the additional AMOST method treatment using acetone as the AMO-solvent  
 4 (b) sample prepared by conventional co-precipitation method in water at pH 10.

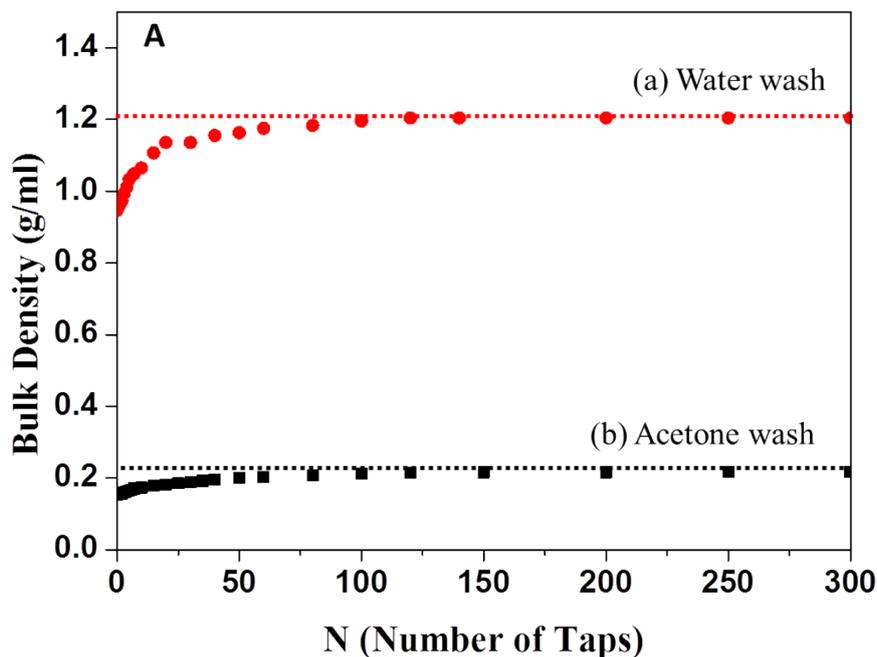
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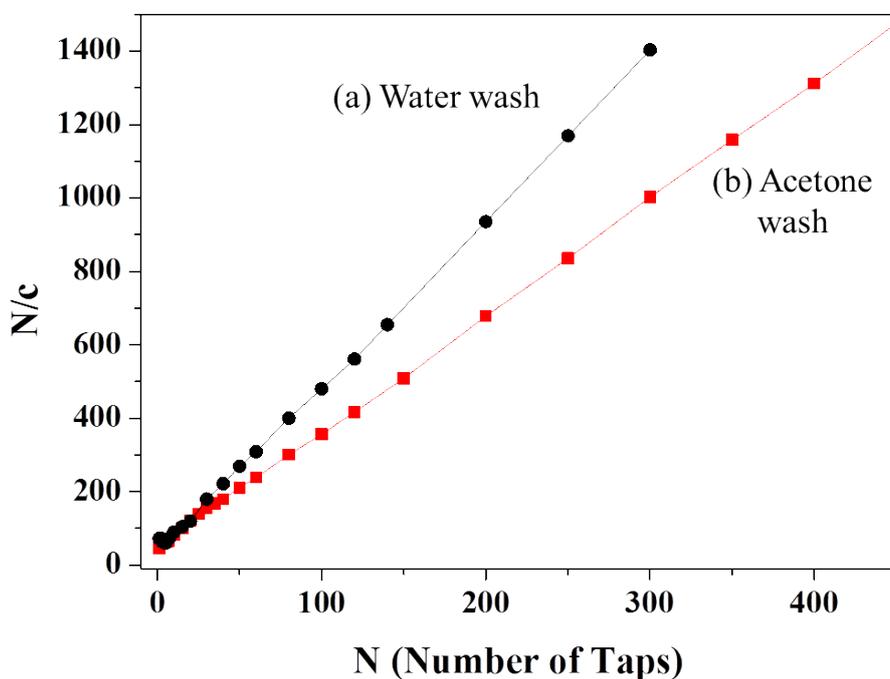
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 8 **Fig. S19** BET Isotherms of  $\text{Mg}_3\text{Al-CO}_3\text{-12}$  (a) sample prepared under identical synthesis  
 9 conditions with the additional AMOST method treatment using methanol as the AMO-  
 10 solvent (b) sample prepared by conventional co-precipitation method in water at pH 12.

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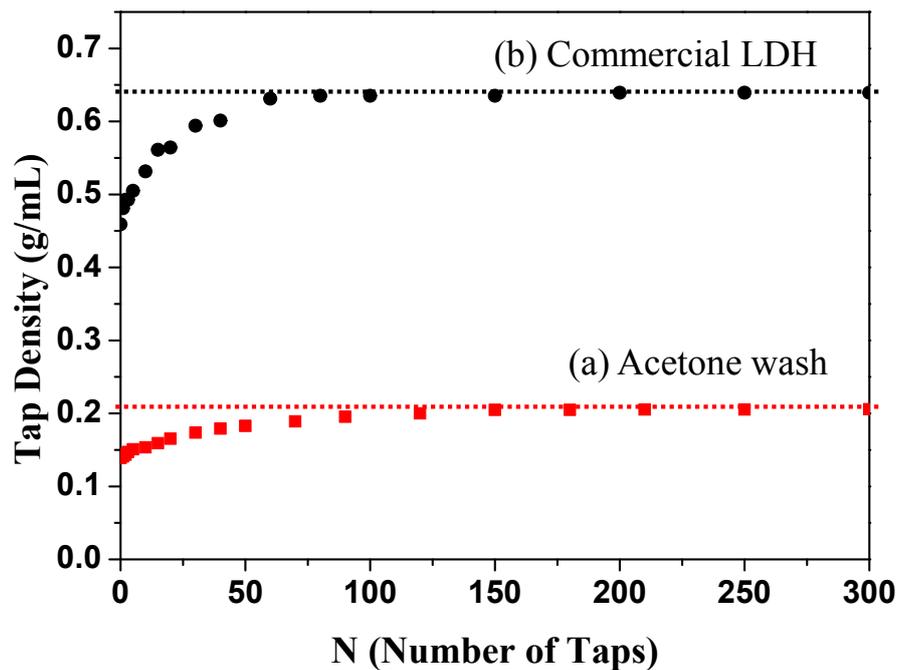
## 1 2.7 Density studies



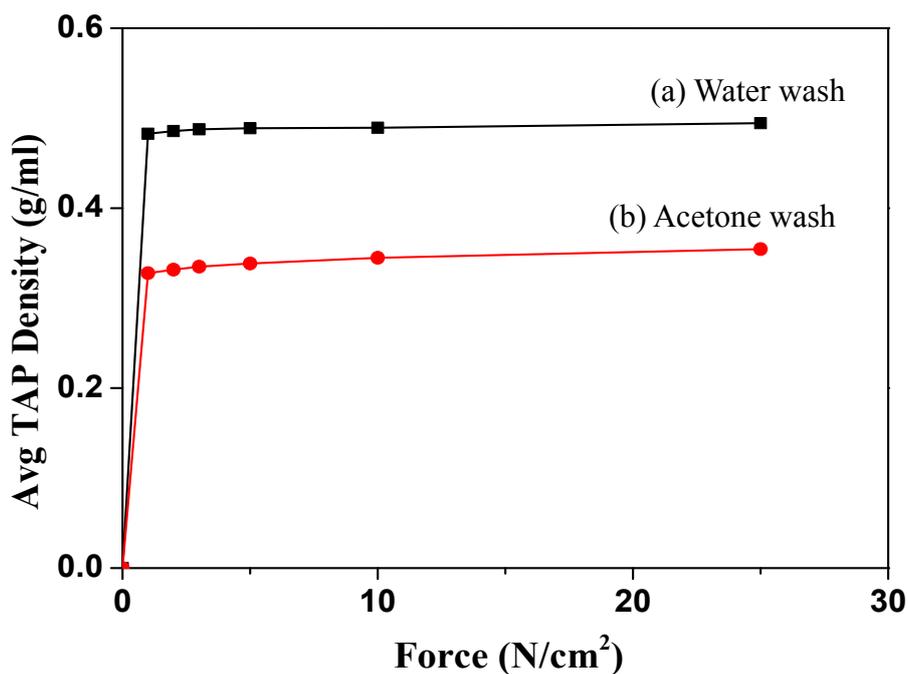
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3 **Fig. S20** Bulk density of Mg<sub>3</sub>Al-NO<sub>3</sub>-10 LDH; (a) sample prepared by a conventional co-  
4 precipitation method in water at pH 10 and (b) AMOST method using the AMO-solvent  
5 acetone.



6  
7 **Fig. S21** Carr's index curves of Mg<sub>3</sub>Al-NO<sub>3</sub>-10 LDH; (a) sample prepared by a conventional  
8 co-precipitation method in water at pH 10 and (b) AMOST method using the AMO-solvent  
9 acetone.



1  
 2 **Fig. S22** Bulk density of  $Mg_3Al-CO_3-10$  LDH; (a) sample prepared by a conventional co-  
 3 precipitation method in water at pH 10 and (b) AMOST method using the AMO-solvent  
 4 acetone.



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 6 **Fig. S23** GeoPyc T.A.P density of  $Mg_3Al-CO_3-10$  LDH; (a) sample prepared by a  
 7 conventional co-precipitation method in water at pH 10 and (b) AMOST method using the  
 8 AMO-solvent acetone.

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 10  
 11

1 [1] Q. Wang and D. O'Hare, *Chemical Communications* 2013, **49**, 6301-6303.