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1 Experimental details

1.1 The synthesis of Mg$_2$Al-CO$_3$-10 LDHs

The metal precursor solution (50 mL) of 0.75 M Mg(NO$_3$)$_2$·6H$_2$O and 0.375 M Al(NO$_3$)$_3$·9H$_2$O was added drop-wise into the 50 mL of 0.5 M Na$_2$CO$_3$ 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$_2$Al-CO$_3$-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$_2$Al-CO$_3$-10-A.

1.2 The synthesis of Mg$_3$Al-CO$_3$-12 LDHs

The metal precursor solution (50 mL) of 0.75 M Mg(NO$_3$)$_2$·6H$_2$O and 0.25 M Al(NO$_3$)$_3$·9H$_2$O was added drop-wise into the 50 mL of 0.5 M Na$_2$CO$_3$ 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$_3$Al-CO$_3$-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$_3$Al-CO$_3$-12-A. When method was used as AMO solvent, the final sample name is Mg$_3$Al-CO$_3$-12-M.

1.3 The synthesis of Mg$_3$Al-NO$_3$-10 LDHs

The metal precursor solution (50 mL) of 0.75 M Mg(NO$_3$)$_2$·6H$_2$O and 0.25 M Al(NO$_3$)$_3$·9H$_2$O was added drop-wise into the 50 mL of 0.5 M NaNO$_3$ 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$_2$ gas to prevent the contamination by atmospheric CO$_2$. 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$_3$Al-NO$_3$-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$_3$Al-NO$_3$-10-A.

1.4 The synthesis of Mg$_3$Al-SO$_4$-10 LDHs

The metal precursor solution (50 mL) of 0.75 M MgSO$_4$·7H$_2$O and 0.25 M Al$_3$(SO$_4$)$_2$·16H$_2$O was added drop-wise into the 50 mL of 0.5 M Na$_2$SO$_4$ base solution. The pH value was kept at ca. 10.0 by wise-dropping 4.0 M NaOH solution. During the synthesis, the system was purged with N$_2$ gas to prevent the contamination by atmospheric CO$_2$. 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$_3$Al-SO$_4$-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$_3$Al-SO$_4$-10-A.

1.5 The synthesis of Mg$_3$Al-Cl-10 LDHs

The metal precursor solution (50 mL) of 0.75 M MgCl$_2$·6H$_2$O and 0.25 M AlCl$_3$·6H$_2$O was added drop-wise into the 50 mL of 0.5 M NaCl base solution. The pH value was kept at ca. 10.0 by wise-dropping 4.0 M NaOH solution. During the synthesis, the system was purged with N$_2$ gas to prevent the contamination by atmospheric CO$_2$. 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$_3$Al-Cl-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$_3$Al-Cl-10-A.

1.6 The synthesis of Mg$_3$Al$_{0.5}$Fe$_{0.5}$-NO$_3$ -10 LDHs

The metal precursor solution (50 mL) of 0.75 M Mg(NO$_3$)$_2$·6H$_2$O, 0.125 M Al(NO$_3$)$_3$·9H$_2$O and 0.125 M Fe(NO$_3$)$_3$·9H$_2$O was added drop-wise into the 50 mL of 0.5 M NaNO$_3$ base solution. The pH value was kept at ca. 10.0 by wise-dropping 4.0 M NaOH solution. During the synthesis, the system was purged with N$_2$ gas to prevent the contamination by atmospheric CO$_2$. 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$_3$Al$_{0.5}$Fe$_{0.5}$-NO$_3$ -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$_3$Al$_{0.5}$Fe$_{0.5}$-NO$_3$-10-A.

1.7 The synthesis of Zn$_2$Al-Borate-8.3 and Mg$_3$Al-Borate-9 LDHs

The synthesis of Zn$_2$Al-Borate-8.3 and Mg$_3$Al-Borate-9 was according to our previous report\cite{1}. The metal precursor solution (50 mL) of 0.75 M Zn(NO$_3$)$_2$·6H$_2$O and 0.375 M Al(NO$_3$)$_3$·9H$_2$O was added drop-wise into the 50 mL of 1.87 M H$_3$BO$_3$ solution base solution. The pH value was kept at ca. 8.3 by wise-dropping 1.0 M NaOH solution. During the synthesis, the system was purged with N$_2$ gas to prevent the contamination by atmospheric CO$_2$. This nucleation process usually takes 30 min. After aging for 16 h with stirring at 65 °C, 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 Zn$_2$Al-Borate-8.3-10-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 Zn$_2$Al-Borate-8.3-A.

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

2. Characterization

2.1 X-ray diffraction
Fig. S1 XRD patterns of Mg$_2$Al-CO$_3$-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. (*) are Bragg reflections from the Al sample holder.

Fig. S2 XRD patterns of Mg$_3$Al-CO$_3$-12 (a) sample prepared by conventional co-precipitation method in water at pH 12 (b) sample prepared under identical synthesis conditions with the additional AMOST method treatment using acetone as the AMO-solvent.
Fig. S3 XRD patterns of Mg₃Al-SO₄-10 (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. (*) are Bragg reflections from the Al sample holder.

Fig. S4 XRD patterns of Mg₃Al-CO₃-10 (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 methanol as the AMO-solvent. (*) are Bragg reflections from the Al sample holder.
Fig. S5 XRD patterns of Mg$_2$Al-SO$_4$-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 methanol as the AMO-solvent.
2.2 Infrared spectroscopy

**Fig. S6** FTIR patterns of Mg$_3$Al-CO$_3$-12 (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.

**Fig. S7** FTIR patterns of Mg$_3$Al-Cl-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.
Fig. S8 FTIR patterns of Mg$_3$Al-NO$_3$-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.

2.3 Transmission Electron Microscopy

Fig. S9 TEM patterns of Mg$_3$Al-Cl-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.
Fig. S10 TEM patterns of Mg$_3$Al-NO$_3$-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.

Fig. S11 TEM patterns of Mg$_3$Al-SO$_4$-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.

2.4 Scanning Electron Microscopy
**Fig. S12** TEM patterns of Mg$_3$Al-Cl-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.

**Fig. S13** TEM patterns of Mg$_3$Al-NO$_3$-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.

**Fig. S14** TEM patterns of Mg$_3$Al-SO$_4$-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.
**2.5 Thermogravimetric analysis**

Fig. S15. TGA and DTG analysis of Mg$_3$Al-NO$_3$-10 LDHs (a) in the range of 30-600 °C; (b) in the range of 30-230 °C; Mg$_3$Al-NO$_3$-10-W prepared by a conventional co-precipitation method in water at pH 10. Mg$_3$Al-NO$_3$-10-A is prepared by identical conditions in water at pH 10 according to the AMOST method using acetone as the AMO-solvent.
Table S1 Summary of water and AMO-solvent content in the AMO-LDHs compared to conventional C-LDHs as determined by analysis of the TGA data.

<table>
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<tr>
<th>LDH</th>
<th>C-LDH²</th>
<th>AMO-LDH-A¹</th>
<th>AMO-LDH-M¹</th>
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<tr>
<td></td>
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<td>c⁴</td>
<td>b³</td>
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<td>Mg₃Al-CO₃-10</td>
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<td>0.74</td>
<td>0</td>
<td>0.5</td>
</tr>
</tbody>
</table>

¹AMO-LDH-A and AMO-LDH-M are the LDH with the formula of \([M^{z+_{1-x}}M'^{y+x}(OH)_{2}]^{z}(X^{n–})_a/r \cdot bH_2O \cdot c(AMO-solvent)\) (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 = Acetone, M = Methanol).

²C-LDH is an LDH with the formula \([M^{z+_{1-x}}M'^{y+x}(OH)_{2}]^{z}(X^{n–})_a/r \cdot bH_2O\) (2); 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 \(a = z(1-x)+xy–2\).

³b is the water content in the formula (1) and (2).

⁴c is the acetone content in the formula (1).
2.6 BET Analysis

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

**Fig. S17** BET Isotherms of Mg$_3$Al$_{0.5}$Fe$_{0.5}$-CO$_3$-10 (a) sample prepared under identical synthesis conditions with the additional AMOST method treatment using acetone as the AMO-solvent 10 (b) sample prepared by conventional co-precipitation method in water at pH.
Fig. S18 BET Isotherms of Mg$_2$Al-CO$_3$-10 (a) sample prepared under identical synthesis conditions with the additional AMOST method treatment using acetone as the AMO-solvent (b) sample prepared by conventional co-precipitation method in water at pH 10.

Fig. S19 BET Isotherms of Mg$_3$Al-CO$_3$-12 (a) sample prepared under identical synthesis conditions with the additional AMOST method treatment using methanol as the AMO-solvent (b) sample prepared by conventional co-precipitation method in water at pH 12.
2.7 Density studies

Fig. S20 Bulk density of Mg₃Al-NO₃₋₁₀ LDH; (a) sample prepared by a conventional co-precipitation method in water at pH 10 and (b) AMOST method using the AMO-solvent acetone.

Fig. S21 Carr’s index curves of Mg₃Al-NO₃₋₁₀ LDH; (a) sample prepared by a conventional co-precipitation method in water at pH 10 and (b) AMOST method using the AMO-solvent acetone.
Fig. S22 Bulk density of Mg$_3$Al-CO$_3$-10 LDH; (a) sample prepared by a conventional co-precipitation method in water at pH 10 and (b) AMOST method using the AMO-solvent acetone.

Fig. S23 GeoPyc T.A.P density of Mg$_3$Al-CO$_3$-10 LDH; (a) sample prepared by a conventional co-precipitation method in water at pH 10 and (b) AMOST method using the AMO-solvent acetone.