
<table>
<thead>
<tr>
<th>Journal:</th>
<th>Journal of Materials Chemistry A</th>
</tr>
</thead>
<tbody>
<tr>
<td>Manuscript ID:</td>
<td>TA-ART-05-2014-002277.R1</td>
</tr>
<tr>
<td>Article Type:</td>
<td>Paper</td>
</tr>
<tr>
<td>Date Submitted by the Author:</td>
<td>10-Jul-2014</td>
</tr>
</tbody>
</table>
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Cite this: DOI: 10.1039/x0xx00000x

Received 00th January 2012,
Accepted 00th January 2012

DOI: 10.1039/x0xx00000x

www.rsc.org/

1 1. Introduction

Layered double hydroxides (LDHs), also known as brucite-like 2D material, are regarded as an important class of host-guest anionic clays which consists of positively charged metal hydroxide layer as host and negatively charged anionic interlayer as guest.[1] The most common LDH-based materials can be expressed by a general formula \([\text{M}^{x+}\cdot \text{M}^{y+}(\text{OH})_2]^{z}(\text{A}^n)^{x}\cdot \text{mH}_{2}\text{O}\), where \(\text{M}^{x+}\) and \(\text{M}^{y+}\) can be most divalent and trivalent metal cations, respectively; \(x\) is the metal ratio of \(\text{M}^{x+}/\text{M}^{y+}\); \(A\) can be various anions.[2]

Owing to their distinguishing features such as compositional flexibility, high anionic exchange ability and biocompatibility, LDHs have attracted considerable attention in the fields of catalyst, biomedicine, electrochemistry and environmental technologies.[3]

However, the LDHs synthesised by conventional methods are often highly aggregated due to their high charge density and hydrophilicity. As a result, isolated LDH powders exhibit relatively low surface areas and unmodified forms cannot be dispersed in non-polar liquids, this imposes both severe limitations on their ability to be surface functionalised and their practical application. Therefore, in recent years many studies have been performed with the aiming of overcoming this limitation. To date, exfoliation methods and surfactant modification methods have been used to produce individual dispersions of LDH layers or thin nanosheets.[4] The organophilic anions which are used in these exchange processes modify the surface characteristics of LDH nanosheets and so reduce their interaction. However, this method normally requires the use of highly polar or even toxic solvents and a multi-step process. Furthermore, the isolated yields using such methods are relatively low and not practical for scale-up. Xu \emph{et al.} has developed a simple and effective way to obtain individual LDH nanoparticles.[5]

These nanoparticles may be dispersed in aqueous solution. However, we are interested in developing scalable methods for the dispersion of LDHs in non-polar hydrocarbon solvents, this would enable the use of LDHs to be realised in much wider fields of materials chemistry, catalysis and sorption.

Recently, we reported a simple and novel method, called the Aqueous Miscible Organic Solvent Treatment (AMOST) method, to obtain a new generation of Mg-Al borate LDHs which are highly dispersible in non-polar hydrocarbon solvents and exhibit high specific surface area (up to 458.6 m\(^2\) g\(^{-1}\)).[6] In this method, the LDHs were synthesised by conventional co-precipitation method, but the final wet slurry was washed with an aqueous miscible organic (AMO) solvent. We found that the wet form of LDH slurry and 100 % aqueous miscible of solvent (e.g. acetone, methanol) are necessary to help improve the compatibility of LDHs with non-polar hydrocarbon solvents. AMO solvent treatment of LDHs can often lead to dispersion into thin nanosheets or exfoliation to even single layers. If we could apply this simple and cost-
In this work, the AMOST method has been systematically investigated in order to develop a more and comprehensive understanding of the utility of the approach as a general method to prepare AMO-LDHs. Many new general distinguishing features of AMO-LDHs have been discovered by varying the composition of LDHs, the pH of synthesis process and by varying the AMO solvents. We also report a more complete study of their properties and characteristics. In particular, we are now able to define a generalised formula for the AMO-LDH family.

2. Experimental Part

2.1 Materials

16 Mg(NO_3)_2·6H_2O (AR), Al(NO_3)_3·9H_2O (AR), NaOH (AR),
17 Na_2CO_3 (AR), MgCl_2·6H_2O (AR), AlCl_3·6H_2O (AR),
18 NaCl (AR), Fe(NO_3)_3·9H_2O (AR), MgSO_4·7H_2O (AR),
19 Al(NO_3)_3·16H_2O (AR), Zn(NO_3)_2·6H_2O (AR),
20 NaNO_3 (AR), Na_2SO_4 (AR), H_3BO_3 (AR), acetone (99.8%)
21 methanol (99.8%), were purchased from Sigma-Aldrich Co.
22 LLC, and used without further purification.

2.2 Experiments

2.2.1 General synthesis method for AMO-LDH

A metal precursor solution was added drop-wise into a base solution under rapid stirring. During this nucleation step, the pH value was constantly controlled by adding drop-wise a NaOH solution. After aging for 16 hours, the precipitate was washed with DI water until the pH was close to 7. Then, the obtained wet cake solid was dispersed into acetone followed by stirring for 1 - 2 hours. At the end of this dispersion step, the resultant solid was filtered and washed thoroughly with acetone. The final product was dried overnight in a vacuum oven at room temperature.

2.2.2 Specific examples of a conventional LDH (MgAl-CO_3 LDH)

The metal precursor solution (50 mL) of 0.75 M Mg(NO_3)_2·6H_2O and 0.25 M Al(NO_3)_3·9H_2O was added drop-wise into the 50 mL of 0.5 M Na_2CO_3 base solution. The pH value was kept at ca. 10.0 by drop wise addition of a 4.0 M NaOH solution. This nucleation process 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. The final product was dried overnight in a vacuum oven at room temperature. This sample was named as Mg_2Al-CO_3·10-W.

2.2.4 Specific examples of AMO-LDH (MgAl-CO_3 LDH)

In the AMOST method, all the nucleation and aging steps are the same as those in the conventional method described above. The key difference is that in the AMOST method, the wet cake solid LDH obtained after washing with DI water is then further rinsed with acetone thoroughly. This acetone washed LDH is then dispersed in 200 mL acetone to give a slurry and stirred at room temperature for 1 - 2 h. The AMO-LDH is isolated by filtration. Finally, the product is dried overnight in a vacuum oven at room temperature. This sample is named as Mg_3Al-CO_3·10-A.

2.3 Powder X-ray diffraction

Powder X-ray diffraction (XRD) data were collected on a PANalytical X’Pert Pro diffractometer in reflection mode at 40 kV and 40 mA using Cu Kα radiation (α1 = 1.54057 Å, α2 = 1.54433 Å, weighted average = 1.54178 Å). Scans were recorded from 3° ≤ θ ≤ 70° with varying scan speeds and slit sizes.

2.4 Thermogravimetric analysis

Thermogravimetric analysis (TGA) measurements were collected using a Netzsch STA 409 PC instrument. TGA was used to determine the mass loss of a sample as a function of temperature. Approximately 20 mg of sample was heated in a corundum crucible between 30 °C and 600 °C at a heating rate of 10 °C min^{-1} under a stream of compressed N_2 flowing at 50 cm^3 min^{-1}.

2.5 Fourier Transform Infrared spectroscopy

Fourier Transform Infrared (FTIR) spectra were recorded on a Biorad FTS 6000 FTIR Spectrometer. It is equipped with a high performance DuraSampIR II diamond accessory of attenuated total reflection (ATR) mode in the range of 400-4000 cm^{-1} with 100 scans at 4 cm^{-1} resolution. The strong absorptions in the range 1667-2500 cm^{-1} are from the DuraSampIR II diamond surface.

2.6 Transmission Electron Microscopy

Transmission Electron Microscopy (TEM) analysis was performed on a JEOL 2100 microscope with an accelerating voltage of 200 kV. LDH nanoparticles were dispersed in ethanol with sonication and then casted onto copper grids coated with Formvar film.

2.7 Scanning Electron Microscopy

Scanning Electron Microscopy (SEM) analysis was performed on a Zeiss Supra 55 scanning electron microscope with an accelerating voltage of 20 kV. LDH powders were spread on carbon tape adhered to an SEM stage. Before imaging, the samples were coated with a thin platinum layer to prevent charging and to improve the image quality.
2.8 Energy-dispersive X-ray spectroscopy

Energy-dispersive X-ray Spectroscopy (EDX) was used to analyse the ratios of metal ions in the LDHs. LDH powders were spread on the carbon tape and coated with a thin platinum layer. The EDX data were collected from JSM-6610LV low vacuum SEM with an accelerating voltage of 20 kV.

2.9 Surface Area and Pore Size Analysis

Specific surface areas and pore size were analyzed using the Brunauer–Emmett–Teller (BET) method. The samples were measured from the N₂ adsorption and desorption isotherms at 77 K collected from a Quantachrome Autosorb-6 surface area and pore size analyzer. Before each measurement, LDH samples were first degassed overnight at 110 °C.

2.10 Density

The bulk density and tap density of AMO-LDHs and C-LDHs were measured by both standard test method (ASTM D7481-09) and GeoPyc 1360 Envelope Density Analyzer in Micrometric company. The procedure for standard test could be described as follows: The powder freely flowed into a graduated cylinder (10 mL) via the funnel. The cylinder with powder was tapped at an interval time of 30 s and fell at the constant height of 20 mm for each tap. The volume was measured before and after 300 taps. The Loose Bulk Density and Tap Density can be calculated by the equation (1) and (2), respectively.

\[
\text{Loose Bulk Density} = \frac{m}{V_0}
\]

\[
\text{Tap Density} = \frac{m}{V_f}
\]

Where \( m \) is the powder weight in the graduated cylinder, \( V_0 \) is the initial powder volume in the cylinder before tapping, \( V_f \) is the final powder volume in the cylinder after 300 taps.

Table 1

<table>
<thead>
<tr>
<th>Property</th>
<th>Parameters</th>
<th>AMO-LDH</th>
<th>C-LDH</th>
<th>Change</th>
</tr>
</thead>
<tbody>
<tr>
<td>Loose Bulk Density ( ^1 )</td>
<td>(g/mL)</td>
<td>0.1-0.18</td>
<td>0.29-0.95</td>
<td>38-84 %</td>
</tr>
<tr>
<td>Tap Density ( ^2 )</td>
<td>(g/mL)</td>
<td>0.16-0.26</td>
<td>0.39-1.2</td>
<td>41-82 %</td>
</tr>
<tr>
<td>Carr’s Index ( ^3 )</td>
<td></td>
<td>32-40</td>
<td>22-39</td>
<td>0-45 %</td>
</tr>
<tr>
<td>Avg TAP Density ( ^4 )</td>
<td>(g/mL)</td>
<td>0.35</td>
<td>0.49</td>
<td>39%</td>
</tr>
<tr>
<td>Surface Pore Volume ( ^5 )</td>
<td>(m²/g)</td>
<td>61-301</td>
<td>1-148</td>
<td>34-11,100 %</td>
</tr>
<tr>
<td>Thermal Changes ( ^6 )</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>T1(°C)</td>
<td></td>
<td>150-191</td>
<td>169-205</td>
<td>-6 - -9 (°C)</td>
</tr>
<tr>
<td>T2(°C)</td>
<td></td>
<td>340-392</td>
<td>392-424</td>
<td>-7 - -8 (°C)</td>
</tr>
</tbody>
</table>

2.11 Structural properties

The structures of the obtained AMO-LDHs and C-LDHs were analyzed using X-ray diffraction (Fig 1 and Fig. S1-S8). The results show that all AMO-LDHs exhibit the same XRD patterns as those of the conventional LDHs, indicating that the AMOST treatment does not affect the structure of the LDHs.

3. Results and discussion

3.1 Physical properties of AMO-LDHs

Aqueous miscible organic-layered double hydroxide (AMO-LDHs) have been synthesised and their properties with respect to structure, morphology, surface, thermal and packed density have been studied. Some of the distinguishing features are listed in Table 1. Generally, AMO-LDHs exhibit lower packed powder density, higher surface area and lower decomposition temperature comparing to conventional layered double hydroxide (C-LDHs).

Fig. 1 XRD patterns for 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 acetone as the AMO-solvent. (*) are Bragg reflections from the Al sample holder.

For Mg₃Al-CO₃ LDH, acetone washed (AMO-LDH) and the conventional water washed (CvLDH), the expected 00l (#l = 3, 6,
9) Bragg reflections were observed (Fig. 1). This is also observed for Mg$_2$Al$_2$CO$_3$-10 LDH, Mg$_2$Al$_2$CO$_3$-12, Mg$_2$Al$_2$SO$_4$-10, Mg$_2$Al$_2$CO$_3$-10-M, and Mg$_2$Al$_2$SO$_4$-10-M (see Figs S1 - S5 respectively).

Similarly, the FTIR spectrum of Mg$_2$Al$_2$CO$_3$-10 LDH shows that the structure of the LDH is unchanged using both synthesis methods (Fig. 2). Similar results were observed for Mg$_2$Al$_2$CO$_3$-12, Mg$_2$Al$_2$Cl$_2$-10, and Mg$_2$Al$_2$NO$_3$-10 (see Figs. S6-S8 respectively).

Fig. 2 shows the FTIR spectrum for the AMO- and C-LDH. Both samples show the characteristic bands for the intercalated CO$_3^{2-}$ ($\nu = 1368$ cm$^{-1}$). Similar bands are observed when the LDHs were synthesised at pH = 12 (Fig. S6). When MgAl$_2$NO$_3$-10 LDHs (acetone or water washed) were synthesised, the band corresponding to the anion was shown at $\nu = 1350$ cm$^{-1}$ (Fig. S8).

3.3 Morphological study

The morphologies of LDHs were examined by TEM. Owing to the relatively high surface charge and hydrophilic nature of LDHs, we find that these particles or crystallites of conventionally synthesised C-LDHs are generally highly aggregated. The Mg$_2$Al$_2$CO$_3$-10-W particles in Fig. 3a are stone-like. On the other hand, the TEM image in Fig. 3b for Mg$_2$Al$_2$CO$_3$-10-A LDH sample shows a flower-type sample. The diameter of the flower is 120 - 250 nm. Similar results were observed for Mg$_2$Al$_2$Cl$_2$-10, Mg$_2$Al$_2$NO$_3$-10 and Mg$_2$Al$_2$SO$_4$-10 (see Fig. S9-S11 respectively). The morphologies of the synthesised LDH powders were studied by SEM. The SEM of Mg$_2$Al$_2$CO$_3$-10 is shown in Fig. 4. It can be easily observed that the Mg$_2$Al$_2$CO$_3$-10-W platelet particles (Fig. 4a) are densely stacked on their $ab$ face, resulting in the formation of hard stone-like clot (Fig. 4a inset) with a smooth surface. For Mg$_2$Al$_2$CO$_3$-10-A (Fig. 4b), most particles are around 150 nm in size and stack in the c-direction to form loose flower-shape agglomerates, which have a more exposed surface. The same results were also found for other LDHs as shown in Fig. S12-S14. A commercial LDH (PURAL MG 62 HT) was also studied. The SEM image of a sample of PURAL MG 62 HT is shown in Fig. 4c, the sample is composed of agglomerates of around 25 µm in size.
Fig. 4 SEM images for Mg₃Al₃CO₃v₁₀; (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 (inset: the images at lower magnification); (c) a sample of a commercial hydrotalcite (PURAL MG 62 HT, from Sasol Ltd).

3.4 Thermal properties

The thermal properties of AMO-LDHs and C-LDHs were investigated using thermogravimetric analysis (TGA). As shown in Fig. 5a and Fig. S15a, the TGA curves of both AMO-LDH and C-LDHs exhibit the typical thermal decomposition behaviour of LDHs which has three weight losses in the range of 50 – 800 °C. By differentiation of the TGA curves, more detailed information regarding the sublety of the thermal behaviour can be obtained as shown in DTG curves (Fig. 5a). It can be clearly found that all LDHs have two distinct events around 200 °C (noted as T1) and 400 °C (noted as T2). The weight loss below T1 is due to the desorption of physisorbed and intercalated solvents. After T1, the hydroxyl groups start to decompose and gradually transform the LDH structure. This reaches a maximum at around 400 °C (T2), and is normally ascribed to the partial decomposition of carbonate anions and complete dehydroxylation of the metal hydroxide layers. However, the AMO-LDHs (either acetone or methanol) show lower decomposition temperatures compared to the C-LDHs. We believe this is a significant difference and is mainly due to the better dispersion of the AMO-LDH particles.

Table 2 highlights the changes in T1 and T2 for all the AMO-Mg₃Al₃CO₃v₉ (x = 2 or 3) compared with their equivalent C-LDH. For T1, the differences range from 6 to 19 °C lower for the AMO-LDHs compared to the corresponding C-LDHs. The change in T2 is more dramatic, with a decrease of 84 °C from 424 °C (for Mg₃Al₃CO₃-10-W) to 340 °C (for Mg₃Al₃CO₃-10-A).

Furthermore, we find an additional reproducible feature in the TGA of AMO-LDHs; this mass loss event is observed at low temperature (30 – 230 °C) as shown in Fig. 5b. We believe this mass loss is due to desorption of the AMO solvents from the AMO-LDHs, which have lower boiling points than H₂O. We find this feature in all AMO-LDHs such as Mg₃Al₃NO₃-10-A, Mg₃Al₃SO₄-10-A, Mg₃Al₃Fe₀.₅CO₃-10-A (see Fig. S15). This signature enable us to have define a new compositional formula for this family: [Mₙ⁺ₐ₋ₓMₗ⁺ₓ(OH)₂]ₓ(ₐ₋ₓ)bH₂O•c(AMO-solvent), which instantly distinguishes them from the normal general formula of LDH [Mₙ⁺ₐ₋ₓMₗ⁺ₓ(OH)₂]ₓ(ₐ₋ₓ)bH₂O, 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 is 1 to 3 and a = z(1– x)+y–2. The details of the composition determined for each AMO-LDH and C-LDH are listed in Table 3 (Table S1). We find that AMO solvent content (c) in these samples is typically in the range of 0.04-0.18, which is 7 - 28 % in total solvent present (water and AMO-solvent). The AMO solvent is probably both bound to the surface of LDH and/or intercalated in the galleries of the LDH. Due to low content and small molecular size of these solvents (0.13 nm for methanol[10] and 0.44 nm for acetone[11]) compared to the main interlayer anions (such as CO₃²⁻, 0.507 nm[12]), the XRD patterns do not resolve any difference between an AMO-LDH and a C-LDH.
1.5 Surface analysis

We were particularly interested in the surface area and pore volume of the AMO-LDHs. We recently reported that borate intercalated AMO-LDHs exhibit surface areas in excess of 301 m$^2$/g (Zn$_2$Al-Borate-8.3-A), compared to the N$_2$ BET surface area of 13 m$^2$/g for the equivalent Zn$_2$Al-Borate-8.3-W prepared in water.\(^{(6a)}\) The pore volume of 2.15 cc.g$^{-1}$ for Zn$_2$Al-Borate-8.3-A which is 2534 times higher than the equivalent C-LDH sample. The theoretical maximum surface area for an AMO-LDH could be as high as 850 m$^2$/g if we could achieve fully dispersed nanosheets. In order to determine if these high surface areas is a general phenomena we have extensively investigated the surface properties of a range of LDHs containing different cation and anion compositions. The surface properties are summarised in Table 4, the data clearly show that all AMO-LDHs have much higher surface areas than those of the equivalent C-LDHs. However, the percentage increase in the N$_2$ BET surface area in each case varies from 34 - 26,200 % and the increase in the total pore volume is from 11 - 147,329 % comparing the C-LDH to the AMO-LDH. To date, MgAl-NO$_3$-10-A LDH has demonstrated the greatest increase of 11,167% in surface area, from 1.5 m$^2$/g$^{-1}$ to 169 m$^2$/g$^{-1}$. Furthermore, an increase of 9,581 % was observed in pore volume (0.0066 and 0.639 cc.g$^{-1}$ for C- and AMO-LDH, respectively).

3.6 Density

The powder density of the AMO-LDHs is one of the most striking initial observations upon initial synthesis and drying. We have investigated the powder density of the AMO-LDHs and compared the results with in the C-LDH and some commercial LDHs, using two measurement methods, a ASTM standard test method (ASTM D7481-09) and using a GeoPyc 1360 Envelope Density Analyzer. Fig. 7 shows the density curves of Mg$_3$AlSO$_4$-10 with and without AMO solvent treatment using the ASTM D7481-09 method. The densities of both Mg$_3$AlSO$_4$-10-A and Mg$_3$AlSO$_4$-10-W increase with increasing tap number and reach a steady value after 100 taps. The loose bulk density can be obtained from the initial point and the last equilibrium point, respectively. It can be found that Mg$_3$AlSO$_4$-10-A has a bulk powder density of 0.1 g.mL$^{-1}$ and a tap density of 0.16 g.mL$^{-1}$.
which is much lower than those of Mg\textsubscript{3}Al-SO\textsubscript{4}-10-W (0.41 g.m\textsuperscript{-1} and 0.63 g.m\textsuperscript{-1}, respectively). The photos of both samples (1 g) after 300 taps clearly show that the AMO-LDH is much more loose compared to that using traditional method. This is ascribed to the delamination of LDH into thin nanosheets after AMO solvent treatment. Similar findings can also be obtained from other AMO-LDHs as shown in Fig. S20 - S23. To confirm the reliability of the obtained tap densities, measurements were also performed on a GeoPyc 1360 Envelope Density. Fig. S23 shows one example for Mg\textsubscript{3}Al-CO\textsubscript{3} LDH. Carr’s Index, also called the Compressibility Index, was used to analyse the measured density results. Carr’s Index can be obtained by plotting the \(N/c\) vs \(N\) according to the Kawakita Equation, \(\frac{N}{c} = \frac{N}{a} + \frac{1}{ab}\), where \(N\) is the number of taps, \(c = (V_{fr} - V_{in})/V_{fr}\). \(V_{fr}\) represents the powder volume, \(V_{in}\) is the initial powder volume before tapping, \(a\) is the Carr’s Index\(^{[7,8]}\), \(b\) is the constant related to cohesiveness and shear strength. The data in Fig. 8 clearly shows that the Carr’s Index (1/slope) of Mg\textsubscript{3}Al-SO\textsubscript{4}-10-A is higher than that of Mg\textsubscript{3}Al-SO\textsubscript{4}-10-W, indicating the AMO-LDHs has a higher compressibility compared to C-LDH.

A summary of the density studies data for all the AMO-LDHs, C-LDHs and commercial LDHs are listed in Tables 5,6. We find that all the AMO-LDHs show lower densities (both loose bulk density and tap density) between 38 – 82 % compared to the equivalent C-LDHs. Compared to commercial LDHs, the AMO-LDH densities are 68 – 70 % less than for example PURAL™ Mg 62. In addition, the Carr’s Indexes of AMO-LDHs are generally higher (0 – 45 %) than those of C-LDHs and commercial LDHs.

### Table 2: Summary of the thermal properties of AMO-LDHs and C-LDHs

<table>
<thead>
<tr>
<th>LDH</th>
<th>AMO-LDH-A(^{1})</th>
<th>C-LDH</th>
<th>Change</th>
<th>AMO-LDH-A(^{1})</th>
<th>C-LDH</th>
<th>Change</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mg\textsubscript{3}Al-CO\textsubscript{3}-12</td>
<td>187</td>
<td>205</td>
<td>-18</td>
<td>382</td>
<td>399</td>
<td>-17</td>
</tr>
<tr>
<td>Mg\textsubscript{3}Al-CO\textsubscript{3}-10</td>
<td>150</td>
<td>169</td>
<td>-19</td>
<td>340</td>
<td>424</td>
<td>-84</td>
</tr>
<tr>
<td>Mg\textsubscript{3}Al-SO\textsubscript{4}-10</td>
<td>181</td>
<td>187</td>
<td>-6</td>
<td>347</td>
<td>392</td>
<td>-45</td>
</tr>
<tr>
<td>Mg\textsubscript{3}Al-SO\textsubscript{4}-12(^{M})</td>
<td>191</td>
<td>205</td>
<td>-14</td>
<td>392</td>
<td>399</td>
<td>-7</td>
</tr>
</tbody>
</table>

\(^{1}\)AMO-LDH-S is the LDH with the formula of \([M\textsuperscript{3+},M\textsuperscript{2+},(OH)\textsubscript{2}]\textsuperscript{z} (X\textsuperscript{−}\textsubscript{a})\textsubscript{n}\textsubscript{H}\textsubscript{2}O\textsubsuperscript{c}(AMO-solvent) \((1)\) wherein \(M\) and \(M'\) are metal cations, \(z = 1\) or 2; \(y = 3\) or 4, 0<\(c\)<1, \(b = 0\)-10, \(c = 0\)-10, \(X\) is an anion, \(r = 1 - 3\) and \(a = z(1-x)+xy-2\).

\(^{M}\)C-LDH is an LDH with the formula \([M\textsuperscript{3+},M\textsuperscript{2+},(OH)\textsubscript{2}]\textsuperscript{z} (X\textsuperscript{−}\textsubscript{a})\textsubscript{n}\textsubscript{H}\textsubscript{2}O\) 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, \(r = 1 - 3\) and \(a = z(1-x)+xy-2\).

\(^{\ast}\)T1 and \(^{\ast}\)T2 are defined as the 1\(^{\ast}\) and 2\(^{\ast}\) minima in 1\(^{\ast}\) differential of the TGA, respectively (i.e. when 2\(^{\ast}\) derivative of the TGA is zero).
**Table 3. Summary of the chemical formula of the AMO-LDHs and C-LDHs using the thermal analysis data**

<table>
<thead>
<tr>
<th>LDH</th>
<th>Formula of AMO-LDH-A (^1)</th>
<th>Formula of C-LDH (^2)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mg₆Al₂CO₃·10</td>
<td>Mg₁₇½Al₂₀(OH)₂(CO₃)₁₂·0.34(H₂O)·0.04(Acetone)</td>
<td>Mg₂₅₃Al₂₅(OH)₃₂(CO₃)₁₄·0.41(H₂O)</td>
</tr>
<tr>
<td>Mg₆Al₂CO₃·10</td>
<td>Mg₁₇½Al₂₀(OH)₂(CO₃)₁₂·0.59(H₂O)·0.18(acetone)</td>
<td>Mg₂₅₃Al₂₅(OH)₃₂(CO₃)₁₄·0.7(H₂O)</td>
</tr>
<tr>
<td>Mg₆Al₂CO₃·12</td>
<td>Mg₁₇½Al₂₀(OH)₂₆(CO₃)₁₄·0.43(H₂O)·0.11(Acetone)</td>
<td>Mg₂₅₃Al₂₅(OH)₃₂(CO₃)₁₄·0.7(H₂O)</td>
</tr>
<tr>
<td>Mg₆Al₂Fe₃½NO₃·10</td>
<td>Mg₁₇½Al₂₀Fe₁₁(OH)₂₀(CO₃)₁₂·0.5(H₂O)·0.06(acetone)</td>
<td>Mg₂₅₃Al₂₅Fe₁₁(OH)₂₀(CO₃)₁₂·0.74(H₂O)</td>
</tr>
<tr>
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<td>Mg₁₇½Al₂₀(OH)₂₆(SO₄)₁₂·0.71(H₂O)·0.17(acetone)</td>
<td>Mg₂₅₃Al₂₅(OH)₃₂(SO₄)₁₂·0.77(H₂O)</td>
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<tr>
<td>Mg₆Al₂NO₃·10</td>
<td>Mg₁₇½Al₂₀(OH)₂₆(NO₃)₁₀·0.38(H₂O)·0.12(acetone)</td>
<td>Mg₂₅₃Al₂₅(OH)₃₂(NO₃)₁₀·0.57(H₂O)</td>
</tr>
<tr>
<td>Mg₆Al₂Cl·10</td>
<td>Mg₁₇½Al₂₀(OH)₂₆(Cl)₁₀·0.46(H₂O)·0.04(acetone)</td>
<td>Mg₂₅₃Al₂₅(OH)₃₂(Cl)₁₀·0.58(H₂O)</td>
</tr>
<tr>
<td>Mg₆Al₂Cl·14</td>
<td>Mg₁₇½Al₂₀(OH)₂₆(CO₃)₁₂·0.43(H₂O)·0.11(methanol)</td>
<td>Mg₂₅₃Al₂₅(OH)₃₂(CO₃)₁₂·0.7(H₂O)</td>
</tr>
</tbody>
</table>

\(^1\)AMO-LDH-S is the LDH with the formula of \([M^{x+}M'^{y+}(OH)_z]^+(X^-)_n\)⁺H₂O⁺(AMO-solvent), wherein M and M' are metal cations, \(z = 1 \text{ or } 2; y = 3 \text{ or } 4, 0 < x < 1, b = 0-10, c = 0-10, X \text{ is an anion, } r = 1-3 \text{ and } a = z(1-x)+y-2. \text{ AMO-solvent (A = Acetone, M = Methanol)}

\(^2\)C-LDH is the LDH with the formula \([M^{x+}M'^{y+}(OH)_z]^+(X^-)_n\)⁺H₂O⁺(2) wherein M and M' are metal cations, \(z = 1 \text{ or } 2; y = 3 \text{ or } 4, 0 < x < 1, b = 0-10, c = 0-10, X \text{ is an anion, } r = 1-3 \text{ and } a = z(1-x)+y-2.

**4. Summary and Conclusions**

We report the synthesis and characterisation of a new family of layered double hydroxides entitled Aqueous Miscible Organic Layered Double Hydroxide (AMO-LDH). We have determined that AMO-LDHs have the unique chemical composition \([M^{x+}M'^{y+}(OH)_z]^+(X^-)_n\)⁺H₂O⁺(AMO-solvent). Although the LDHs that are synthesised using the AMOST and conventional methods possess the same structural properties as shown by XRD. The AMO-LDHs possess new physical properties; they
have significantly higher surface areas, pore volumes, together with lower density and higher compressibility than both conventional and commercial LDHs. The role of the AMO-solvent (eg acetone and methanol) is the subject to further theoretical and experiment investigation. Our current thoughts (Fig. 9) are that the AMO solvent replaces the surface bound water from the surface of primary LDH particles and so rendering them hydrophobic rather than hydrophilic. These hydrophobic particles would then have a much-diminished driving force for aggregation to dense agglomerates.

**Fig. 9** The proposed mechanism for the formation of AMO-LDHs using acetone treatment.

We are already finding that AMO-LDHs are good candidates for wide variety of applications such as additives to polymers, sorbents and catalyst supports. These results will be published in due course.

**Acknowledgements**

This work is supported SCG Chemicals, Bangkok, Thailand and by the Fundamental Research Funds for the Central Universities (TD-JC-2013-3), the Program for New Century Excellent Talents in University (NCET-12-0787).

**Notes and references**

1. Chunping Chen and Miaosen Yang contribute equally to the project and are to be considered co-first authors.