Electronic Supplementary Information

Silylation of Layered Double Hydroxides via an Induced Hydrolysis Method

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TG/DTG analysis

The typical LDH shows three mass loss steps, including dehydration, decarbonation and dehydroxylation processes with some overlaps one another. The whole mass loss of Hw-Si (48.7%) is slightly greater than that of Hd-Si (Figures S1a and b). All the separate mass loss stages are different between these two samples. Firstly, only one DTG peak recorded below 200 °C, assigned to water adsorbed on sample surface. The mass loss is 10.9%, close to that of Ht. Secondly, the mass loss of the second stage for Hw-Si (36.5%) is greater than those of Hd-Si (32.2%) and Ht (33.6%). This is due to the loss of alky chains of silane.

The thermal properties of HSw-Si and HG-Si are similar with each other, but apparently different from the raw LDH sample and the two samples with no surfactant. As shown in Figure S2, for the first mass loss stage, there are 7.5% and 10.9% mass loss detected in HSw-Si and HG-Si, respectively, corresponding to less water adsorbed on the sample surface after DS modified. The greater mass loss of HG-Si may due to the surfactant adsorbed on the LDH surface. The second mass loss increases to 38.3% (HSw-Si) and 37.2% (HG-Si). Three steps are recorded in this stage for HG-Si with the central temperatures at 215 °C, 298 °C and 392 °C, respectively. During these temperature regions, the intercalated anions are decomposed (1350 cm⁻¹) together with partly loss of alkyl chains and hydroxyl groups (Figure 8). The similar procedures are also can be observed for HSw-Si, while only a strong mass loss stage recorded (Figure S2a). There are new mass losses recorded when the temperature is raised over 600 °C, which are corresponding to decomposition of sulfate and the further decomposition of mixed metal oxides. On the basis of the differences above, a conclusion can be drawn that the surfactant shows obvious effect on the thermal properties of the silylated products.
Figure S1. TG/DTG curves of H_d-Si (a) and H_w-Si (b).
Figure S2. TG/DTG curves of HSw-Si (a) and HG-Si (b).
Energy-dispersive X-ray analysis (EDX) analysis

The compositions of the samples were shown in Table S1, deduced by EDX analysis (Figure S3). The contents of Si are relatively lower in H\textsubscript{d}-Si and HG-Si. For the H\textsubscript{d}-Si, most of the silanes added into the reaction system are washed away, and only small amount adsorbed on the surface of LDHs. For the HG-Si, the synthesis reaction was conducted in water medium, so the probability for the silylation reaction is less than that in ethanol. The relative proportion of Si for H\textsubscript{w}-Si and HS\textsubscript{w}-Si are close with each other. The water can induce silane to hydrolysis and condensation with the surface –OH of LDHs in ethanol medium, result in an increase of the Si contents in the final products.

Figure S3 The EDX analysis of the silylation products: a. H\textsubscript{d}-Si, b. H\textsubscript{w}-Si, c. HS\textsubscript{w}-Si, and d. HG-Si.
**Table S1.** The atomic proportion of the silylation products

<table>
<thead>
<tr>
<th>Samples</th>
<th>Mg</th>
<th>Al</th>
<th>Si</th>
<th>S</th>
</tr>
</thead>
<tbody>
<tr>
<td>H\textsubscript{d}-Si</td>
<td>11.64</td>
<td>6.29</td>
<td>0.72</td>
<td>–</td>
</tr>
<tr>
<td>H\textsubscript{w}-Si</td>
<td>20.76</td>
<td>11.31</td>
<td>5.73</td>
<td>–</td>
</tr>
<tr>
<td>HS\textsubscript{w}-Si</td>
<td>10.39</td>
<td>5.79</td>
<td>2.35</td>
<td>0.83</td>
</tr>
<tr>
<td>HG-Si</td>
<td>13.12</td>
<td>5.61</td>
<td>0.59</td>
<td>1.75</td>
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</tbody>
</table>