Supporting Information

Analysis methodology

X-ray diffraction (XRD). The oriented samples were prepared by adding 15 mg solid products into 1 mL deionized water, followed by ultrasonic dispersion for 2 min. Then, the suspension was added onto a glass slide dropwise and dried at ambient temperature. ethylene glycol (EG) saturated samples were prepared by treating the oriented samples in a glass desiccator with EG at ambient temperature for 24 h.

Thermogravimetry analyses (TG). TG tests were performed on a Netzsch STA 409PC instrument. About 15 mg of ground sample was heated in a corundum crucible from 30 to 1000°C at a heating rate of 10°C/min under a pure N_2 atmosphere (60 cm^3/min). The differential thermogravimetric (DTG) curve was derived from the TG curve.

Fourier transform infrared spectroscopy (FTIR). FTIR spectra were obtained using a Bruker VERTEX 70 infrared spectrometer. All spectra were collected at room temperature over the range of 400–4000 cm^{-1} with a resolution of 4 cm^{-1} and by using 64 scans.

RESULTS AND DISCUSSION

Mg/Al ratios on crystallinity of products

The dehydroxylation temperature of both Ht and saponite were highly correlated with their crystallinity and aqueous stability, which can be obtained easily from TG-DTG curves (Fig. S2, Talbe S1). After hydrothermal treatment for 4 days, the sample with

Mg/Al = 4 showed the lowest content of impure phases and the least mass loss of dehydration during the first thermal decomposition stage (Fig. S2); Ht product with Mg/Al = 6 showed the lowest dehydroxylation temperature (390°C), followed by the samples with Mg/Al = 4 (427°C) and 2 (430°C) (Fig. S2). Therefore, it was concluded that the hydrotalcite, which was reconstructed under the condition of Mg/Al = 4 was the most stable. With the increase of Mg/Al, the mass losses of Ht dehydroxylation increased, while the dehydroxylation of the saponite gradually decreased (780°C) (Table S1). This confirmed that the initial mixed oxides were gradually converted to saponite via hydrotalcite. The crystallinity of saponite decreased with the increase of Mg/Al ratio, which is consistent with the XRD results. Increasing the reaction time to 15 days, the dehydration mass loss was still the least for the sample with Mg/Al = 4, and the most for the sample with Mg/Al = 6 (Table S1). The dehydroxylation temperatures of hydrotalcite and saponite for all the three series of samples were ca. 560°C and 780°C, respectively. This result indicated that the crystallinities of the two phases were similar under these conditions, and they did not change with the concentrations of cations initially used. Based upon these results, a conclusion was drawn as follows: the lower Mg/Al ratio with higher Al environment was conducive to the crystal growth of saponite but also resulted in the formation of natrodavyne phase.

Fig. S1.

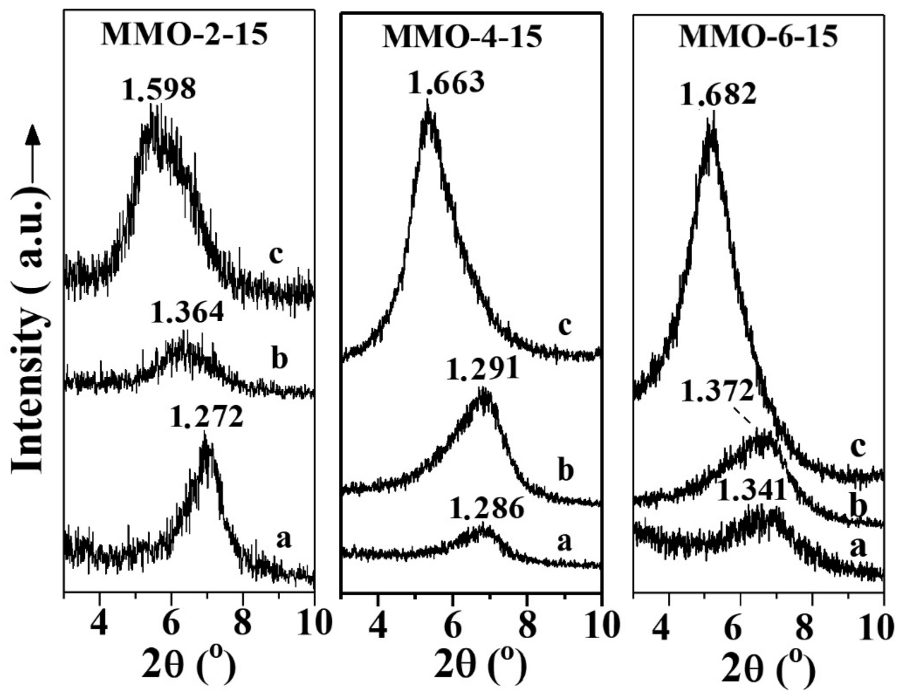


Fig. S1. The XRD patterns of MMO-n-15 products. a. The random samples, b. The orientated samples, and c. The orientated samples with EG saturation.

Fig. S2.

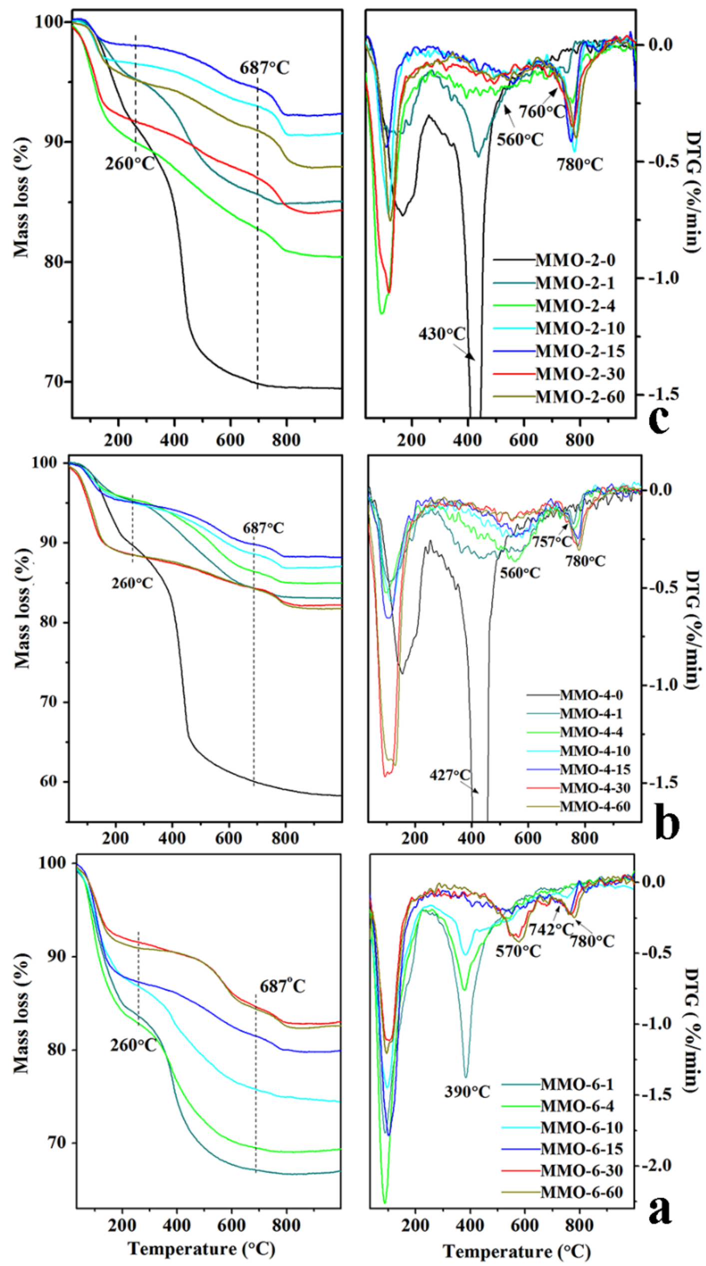


Fig. S2. TG-DTG curves of the hydrothermal treated products. a. MMO-2 series, b. MMO-4 series and MMO-6 series.

Fig. S3.

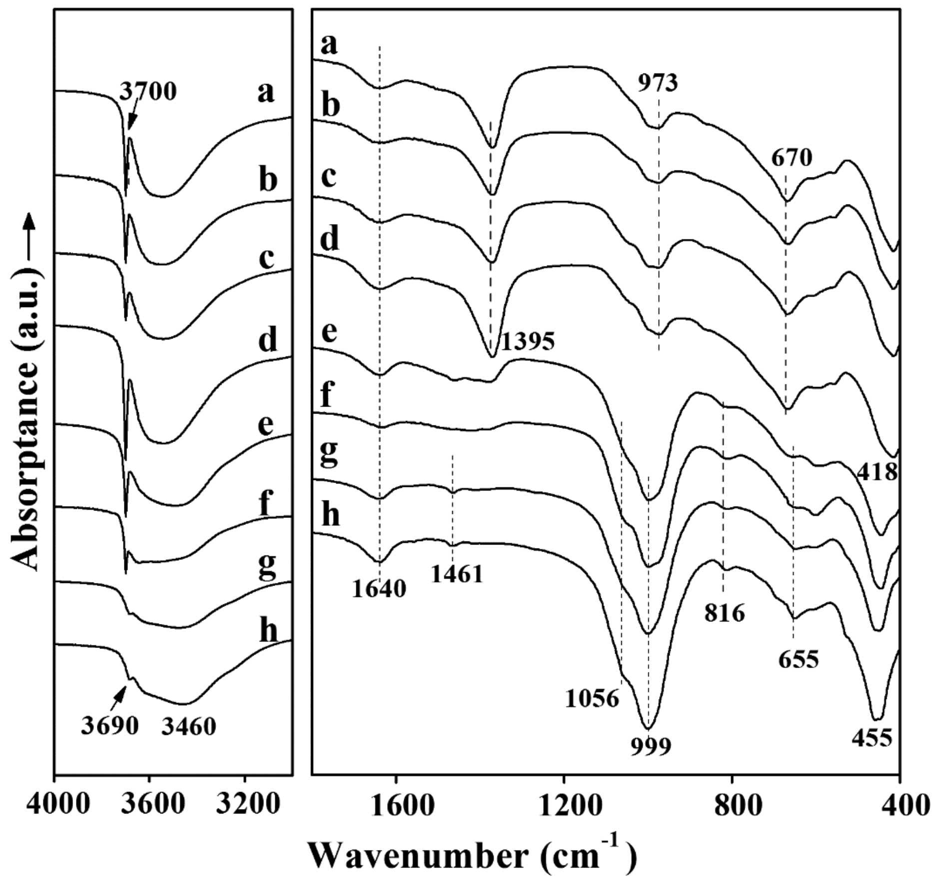
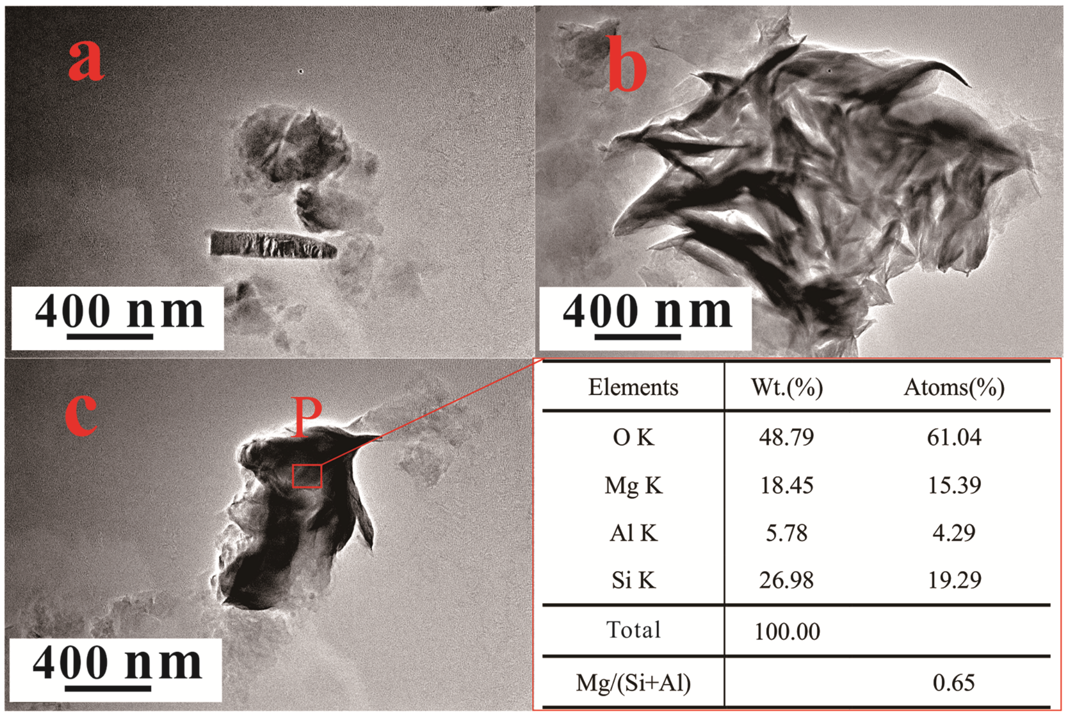


Fig. S3. FTIR spectra of MMO-4 hydrothermal products formed with different SiO_3^{2-} concentrations. a. MMO-4-4-0.5AEC, b. MMO-4-15-0.5AEC, c. MMO-4-4-1.0AEC, d. MMO-4-15-1.0AEC, e. MMO-4-4-4.0AEC, f. MMO-4-15-4.0AEC, g. MMO-4-4-11AEC, and h. MMO-4-15-11AEC.

Fig. S4.



Elements	Wt.(%)	Atoms(%)
O K	48.79	61.04
Mg K	18.45	15.39
Al K	5.78	4.29
Si K	26.98	19.29
Total	100.00	
Mg/(Si+Al)		0.65

Fig. S4. TEM images and EDS results of MMO-2-15. a. the coexistence of saponite and natrodavyne, b. curled saponite flakes, and c. saponite and its EDS results (P).

Fig. S5.

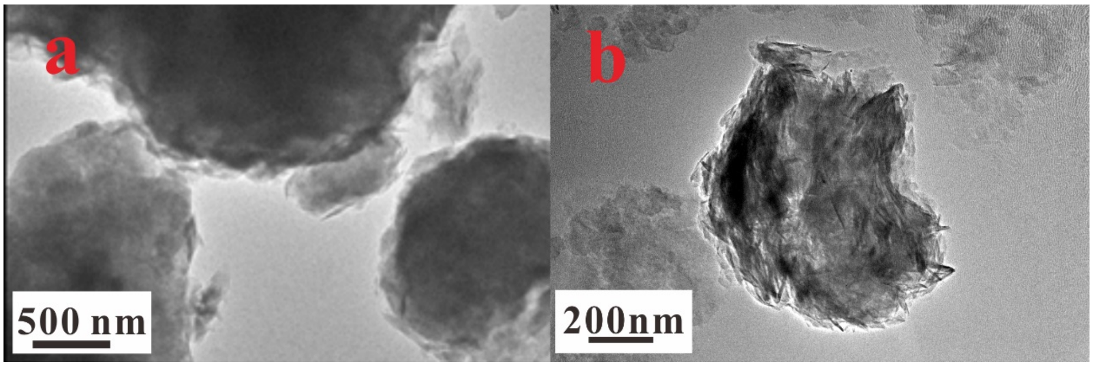


Fig. S5. TEM images of MMO-4 hydrothermal products. a. MMO-4-1, and b. MMO-4-2.

Table S1. Thermal stability of hydrothermal products of MMOs after different reaction times.

Sample	Stage 1		Stage 2		Stage 3	
	t/°C	mass loss/%	t/°C	mass loss/%	t/°C	mass loss/%
MMO-2-0	170	8.66	430	21.45	690	0.28
MMO-2-1	150	4.72	450	9.61	760	0.86
MMO-2-4	100	10.05	430	7.11	760	2.34
MMO-2-10	130	3.43	560	3.44	780	2.51
MMO-2-15	130	1.92	560	3.25	780	2.49
MMO-2-30	150	8.30	560	4.52	780	3.16
MMO-2-60	130`	4.79	560	4.23	780	3.16
MMO-4-0	150	10.28	442	28.59	—	—
MMO-4-1	112	4.75	450	10.99	760	1.21
MMO-4-4	100	4.52	560	9.15	756	1.43
MMO-4-10	100	4.95	541	6.43	772	1.66
MMO-4-15	100	4.87	560	5.26	780	1.63
MMO-4-30	100	11.48	560	4.18	780	2.18
MMO-4-60	100`	11.48	560	4.18	780	2.54
MMO-6-1	100	16.36	444	16.50	760	0.45
MMO-6-4	100	17.17	390	13.28	780	0.49
MMO-6-10	100	13.20	560	11.01	780	1.04
MMO-6-15	100`	12.68	560	5.82	780	1.65
MMO-6-30	100	8.45	560	6.93	780	1.80
MMO-6-60	100`	9.06	560	6.53	780	2.02