Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.


**Supporting information**

**Fig. S1** SEM images of precursors synthesized with different molar ratio of urea to metallic cations (a) 10, (b) 5, (c) 2.5; (d) TG curve of as-obtained precursor-1.

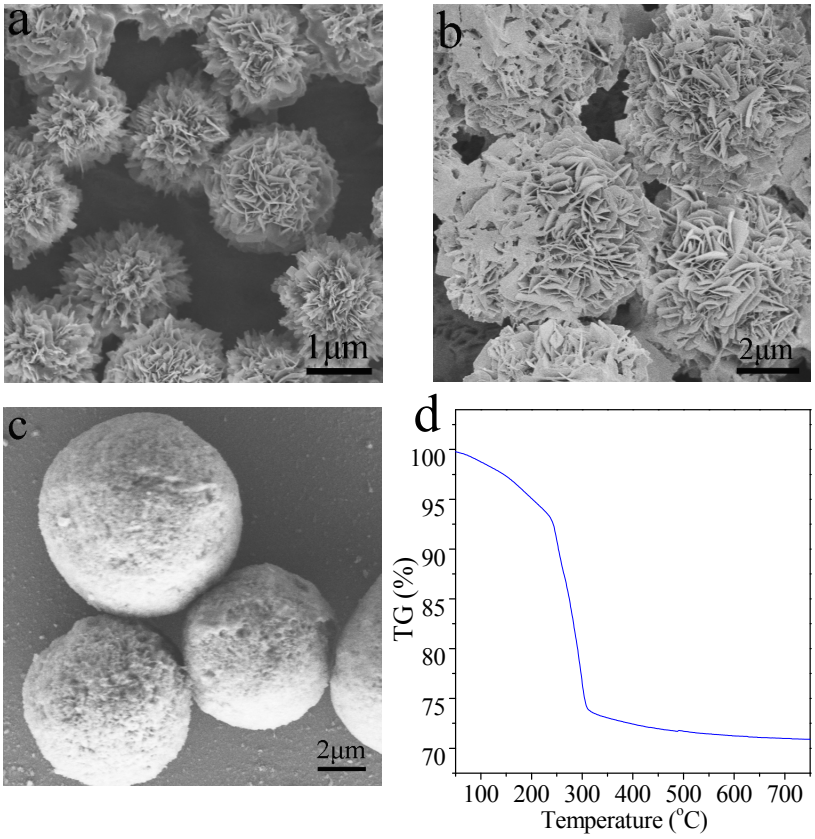

**Fig. S2** XRD patterns of LDH samples synthesized with precursor-1 being calcined at varying temperatures for 4 h.

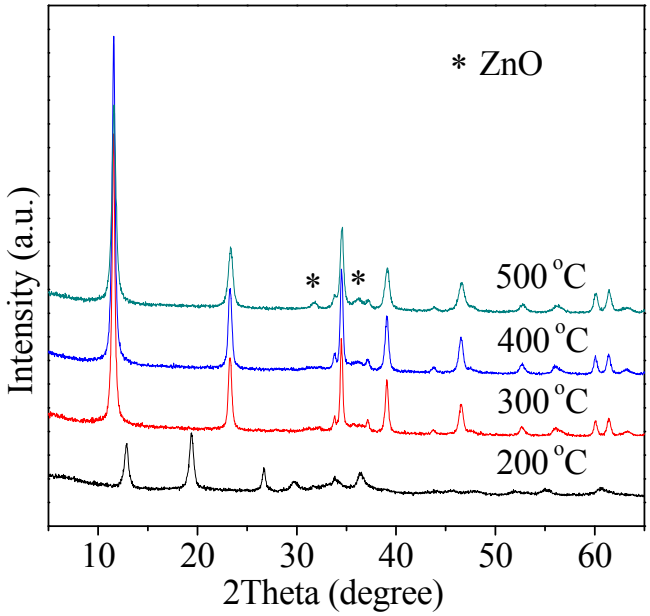

**Fig. S3** SEM images of the precursor-1 calcined at 300 °C with varying heating rate for 4 h: (a, e) 1 °C · $min^{-1}$, (b, f) 5 °C · $min^{-1}$, (c, g) 10 °C · $min^{-1}$, (d, h) 20 °C · $min^{-1}$.

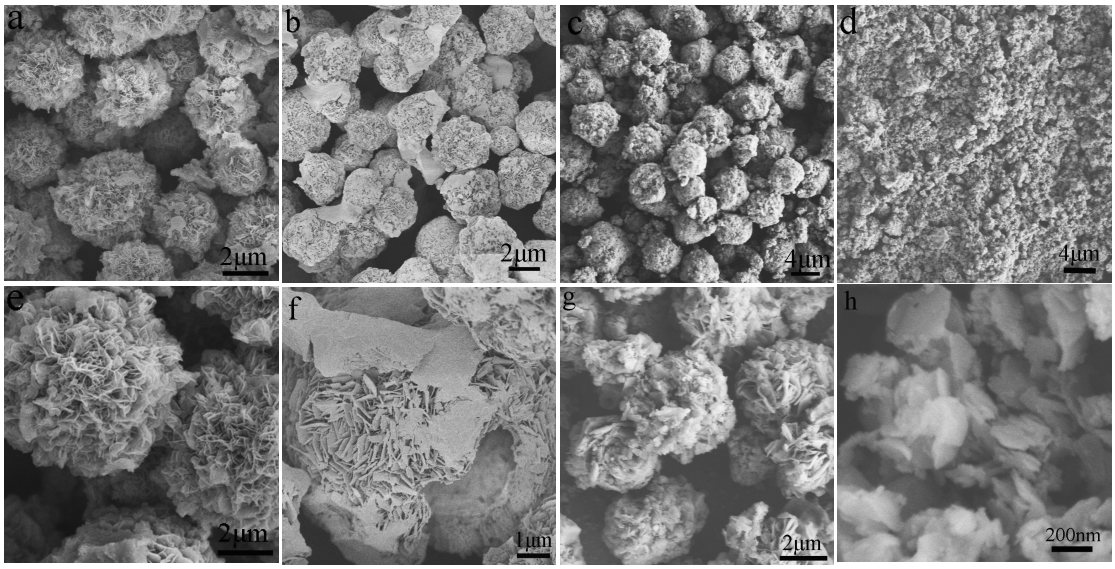

**Fig. S4** XRD patterns of the precipitated mixture synthesized with different reaction time: (a)2 h; (b) 4 h; (c) 8 h; (d) 24 h.

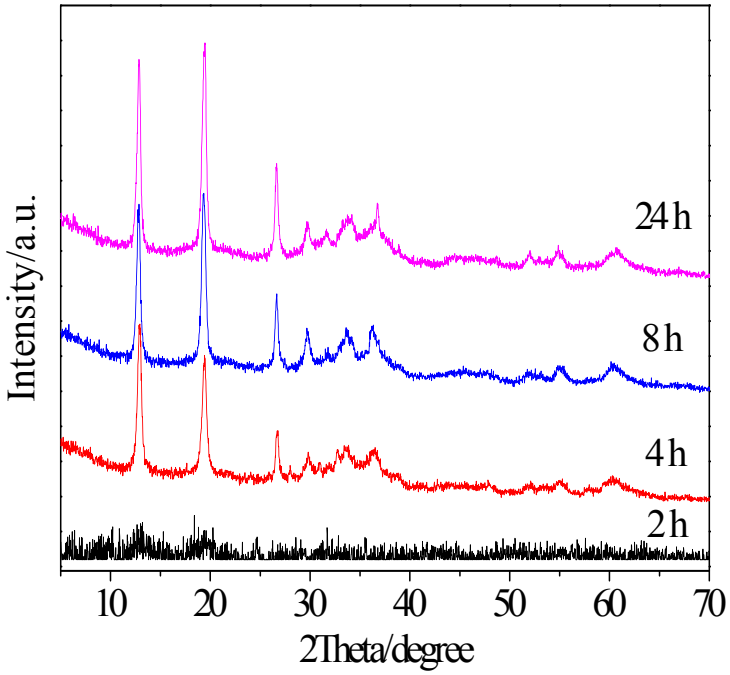

**Fig. S5** XRD pattern and SEM image of sample LDHs synthesized with molar ratio of urea : $(Zn^{2+}+Al^{3+})=1$.

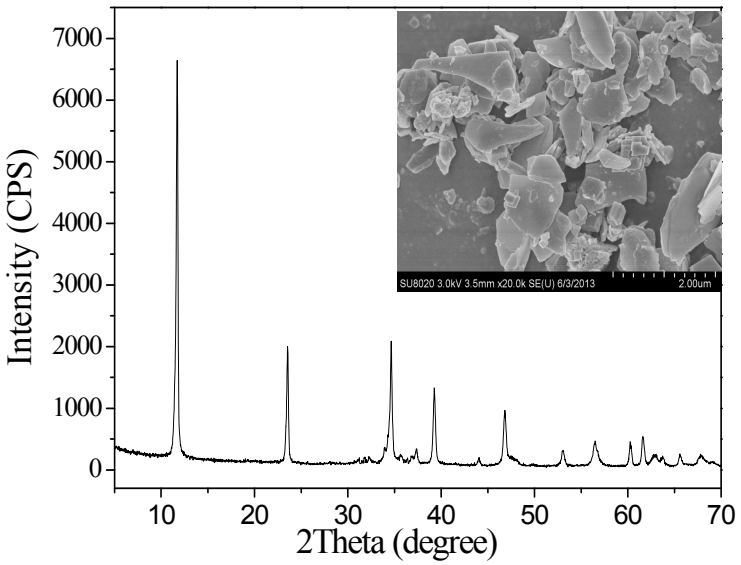

**Fig. S6** Linearized Langmuir adsorption isotherms (a) and Pseudo-second-order kinetics (b) for adsorption of MO onto LDO-1 and LDH-1; (c) XRD patterns of samples obtained with different adsorption time of MO onto LDO-1: (a) 5 min, (b) 30 min, (c) 1 h, (d) 3 h, (e) 8 h, (f) 12 h; (d) FTIR spectra of LDH-1, LDO-1 before and after adsorption of MO.

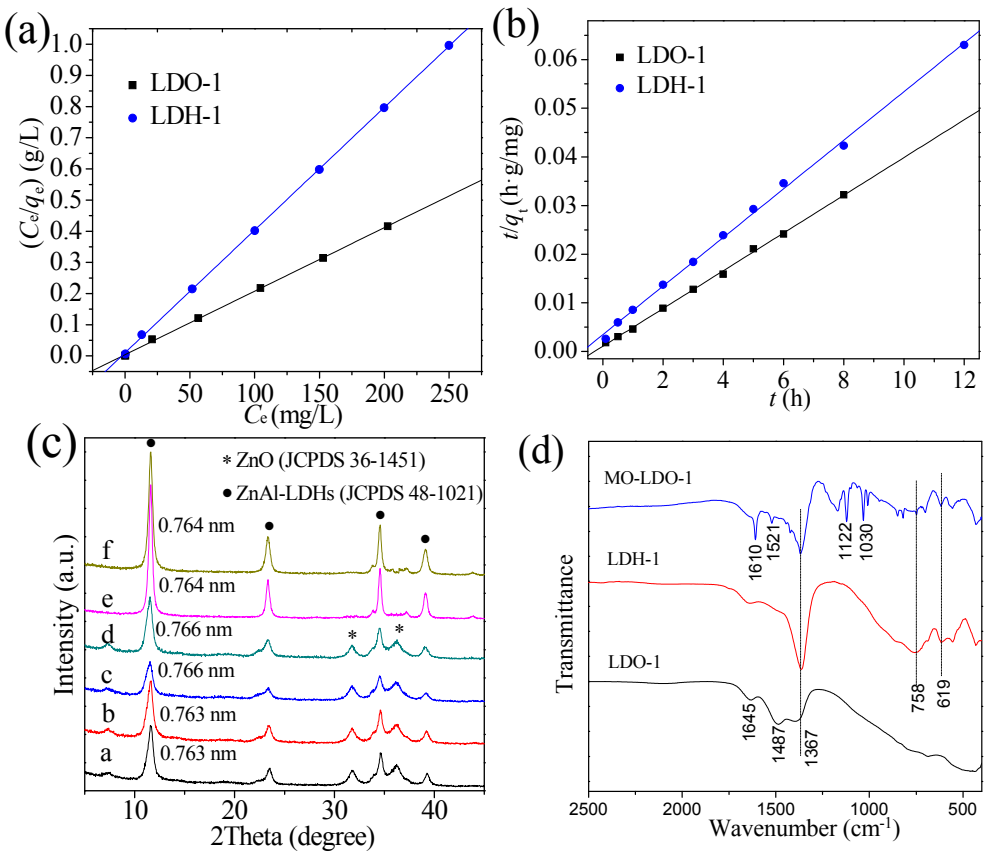

**Table S1** Unit cell parameters and interplanar spacing of ZnAl-LDHs.

| samples | *d*/nm | | | | | *a*/nm | *c*/nm |
|---|---|---|---|---|---|---|---|
| | | 003 | 006 | 110 | | | |
| LDH-1 | 0.760 | 0.380 | 0.154 | 0.308 | 2.281 | | |
| LDH-2 | 0.759 | 0.379 | 0.154 | 0.308 | 2.277 | | |
| LDH-3 | 0.762 | 0.381 | 0.154 | 0.308 | 2.286 | | |

**Table S2** Langmuir and Freundlich model constants for MO adsorption onto the LDH-1 and LDO-1.

| samples | Langmuir | | | Freundich | | |
|---|---|---|---|---|---|---|
| | $q_m$(mg·g$^{-1}$) | $K_L$/(L·mg$^{-1}$) | $R^2$ | $n$ | $K_F$/(L·g$^{-1}$) | $R^2$ |
| LDO-1 | 490.2 | 0.494 | 0.9995 | 4.032 | 179.06 | 0.7982 |
| LDH-1 | 253.8 | 0.392 | 0.9998 | 3.636 | 74.64 | 0.9313 |

**Table S3** Constants of pseudo-first-order kinetic model and the pseudo-second-order kinetic model for adsorption of MO onto LDH-1and LDO-1 samples.

| samples | $q_{e,exp}$/(mg·g$^{-1}$) | Pseudo-second order | | | Pseudo-first order |
|---|---|---|---|---|---|
| | | $q_{e,cal}$/(mg·g$^{-1}$) | $k_2\times10^{-2}$/h$^{-1}$ | $R^2$ | $R^2$ |
| LDO-1 | 248.34 | 263.16 | 1.444 | 0.9985 | 0.9058 |
| LDH-1 | 190.35 | 204.08 | 0.706 | 0.9980 | 0.8824 |