Electronic Supplementary Material (ESI) for RSC Advances.

Supplementary Information

Layered double hydroxide composite monoliths with three-dimensional hierarchical channels: structural control and their adsorption behavior

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Table S1. Influence of base on prepared samples

Reagent	Solution pH			LDHs[c]	State
	0 min[a]	3 min	Final[b]		
PO	1.2	2.6	3.8	yes	Gel
NaOH	1.2	3.9	3.9	yes	Powder
Urea	2.1	2.1	4.6	no	Solution

[a] The compositions of solutions: $AlCl_3 \cdot 6H_2O$, $MgCl_2 \cdot 6H_2O$, water, and ethanol (PO and NaOH systems); $AlCl_3 \cdot 6H_2O$, $MgCl_2 \cdot 6H_2O$, urea, water, and ethanol (urea system)

[b] Measured after 1h (PO and NaOH systems) or 24 h (urea system).

[c] Assessed by XRD analysis.

$AlCl_3 \cdot 6H_2O$ (6.55 mmol) and $MgCl_2 \cdot 6H_2O$ (5.23 mmol) were dissolved in water/ethanol (222/51.4 mmol. PO (26.0 mmol) or NaOH (10 $mol \cdot L^{-1}$, 1.35 mL) was added at 25 °C, and urea (23.8 mmol) was added at 90 °C.

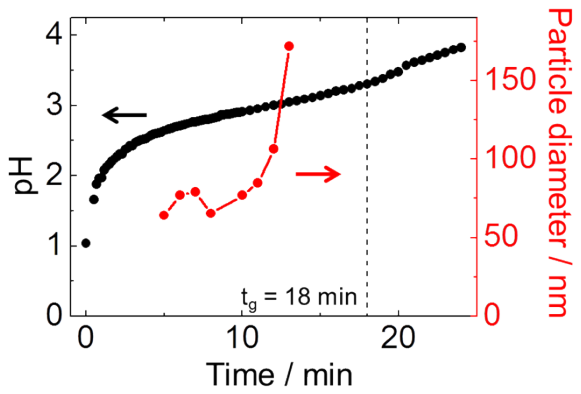

Figure S1. Time evolution of pH and size of particle aggregates in the reaction solution. $AlCl_3 \cdot 6H_2O$ (32.8 mmol) and $MgCl_2 \cdot 6H_2O$ (26.2 mmol) were dissolved in water/ethanol (1.11 mol/257 mmol). PO (131 mmol) was added at 25 °C, and the time evolution of pH in the reaction solution was measured at 40 °C. Black and red circles correspond to pH and particle diameters, respectively. t_g: gelation time. pH at the gelation point was 3.3. Solution pH was measured using a pH meter (HORIBA, Japan). Particle size was measured using a zeta-potential and particle size analyzer (ELSZ-2, Otuka Electronics, Japan).

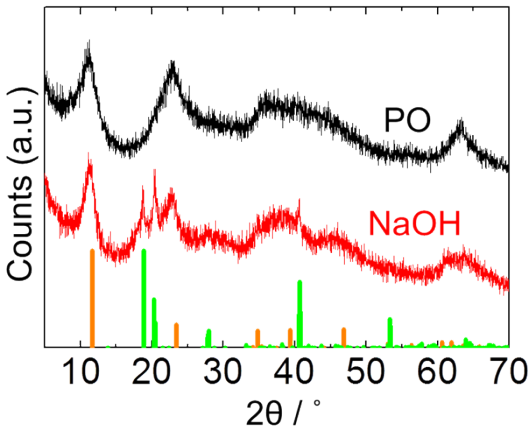

Figure S2. XRD patterns of powder samples obtained from PO and NaOH reactions (yellow: hydrotalcite, green: $Al(OH)_3$, bayerite phase). $AlCl_3 \cdot 6H_2O$ (6.55 mmol) and $MgCl_2 \cdot 6H_2O$ (5.23 mmol) were dissolved in water/ethanol (222/51.4 mmol). PO (26 mmol) or NaOH (10 $mol \cdot L^{-1}$, 1.0 mL) was added at 25 °C and kept for 24 h.

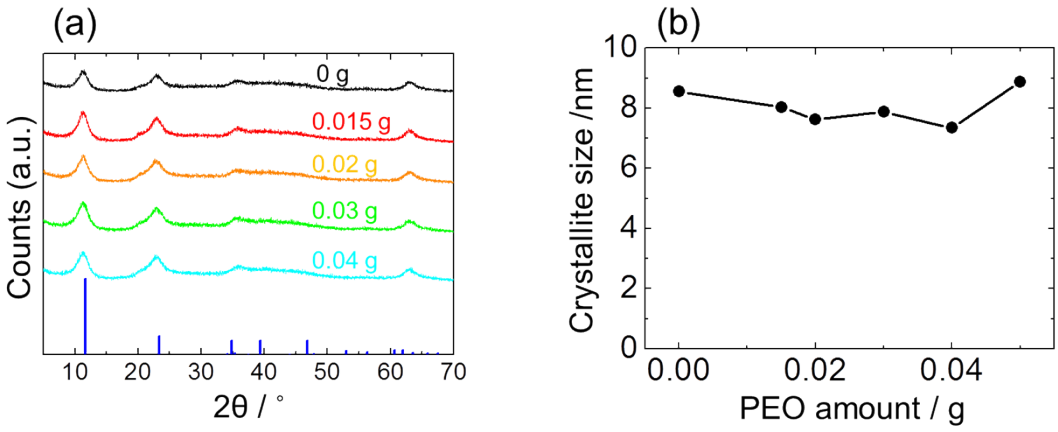

Figure S3. (a) XRD patterns of monolithic xerogels with different macrochannel sizes, resulting from different PEO contents. Blue lines indicate reference $Mg_4Al_2(OH)_{12}CO_3(H_2O)_3$ (JCPDS card no. 70-2151). (b) Relationship between crystallite size and precursor PEO content. Monoliths exhibited similar diffraction patterns, assigned to hydrotalcite-type LDH. Crystallite sizes were estimated from the Scherrer equation, using the LDH (003) reflection of the XRD patterns. Crystallite sizes were independent of PEO content.

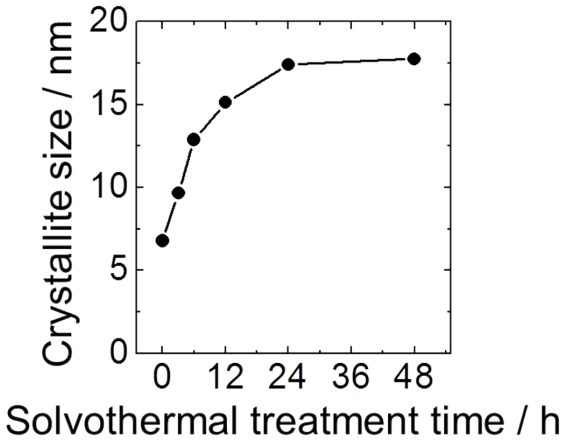

Figure S4. Crystallite size of (003) LDH crystals after solvothermal treatment. Crystallite size of LDH increased with increasing treatment time, from 6.8 to 17.7 nm.

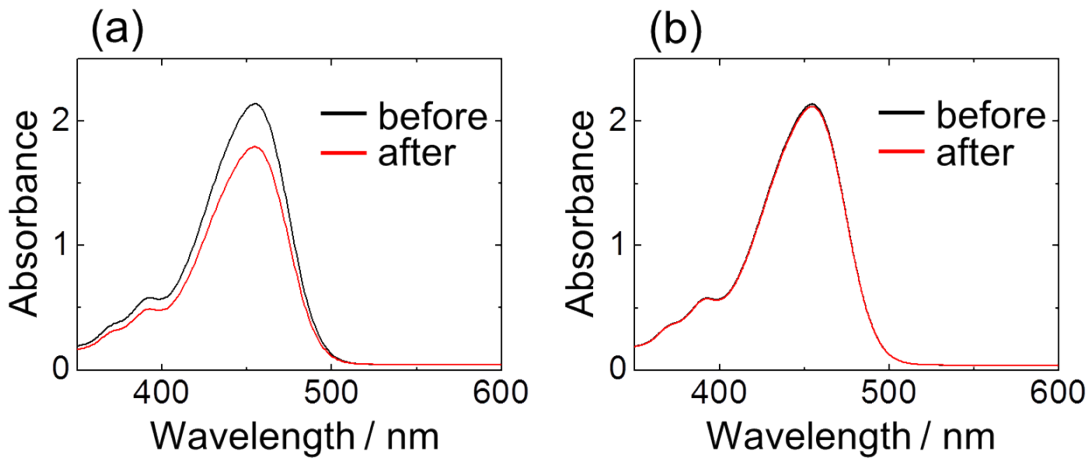

Figure S5. Absorbance spectra of pyranine solution before and after adsorption by (a) LDHs-based monolith and (b) $Al(OH)_3$ powder. 0.06 g of LDH-based monolith and $Al(OH)_3$ (gibbsite) were dispersed in 10 mL of pyranine solution (19.1 mmol/L). After 72 h, supernatants were analyzed by UV-vis absorption spectroscopy. The amount of pyranine adsorbed on LDH was estimated from the Beer-Lambert law to be (a) 17 and (b) <1 %.