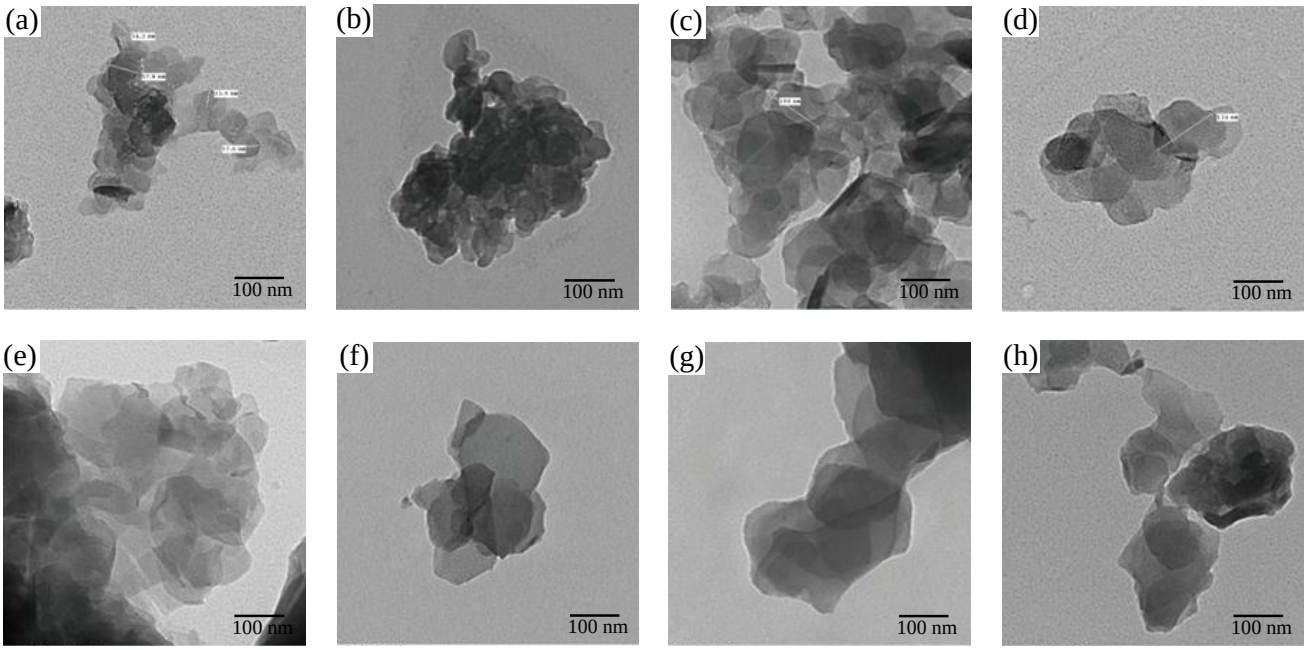
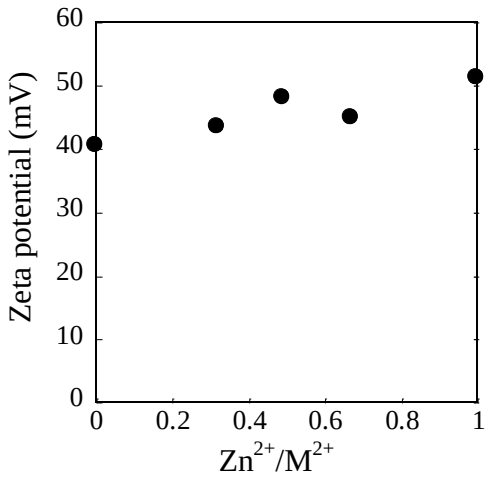
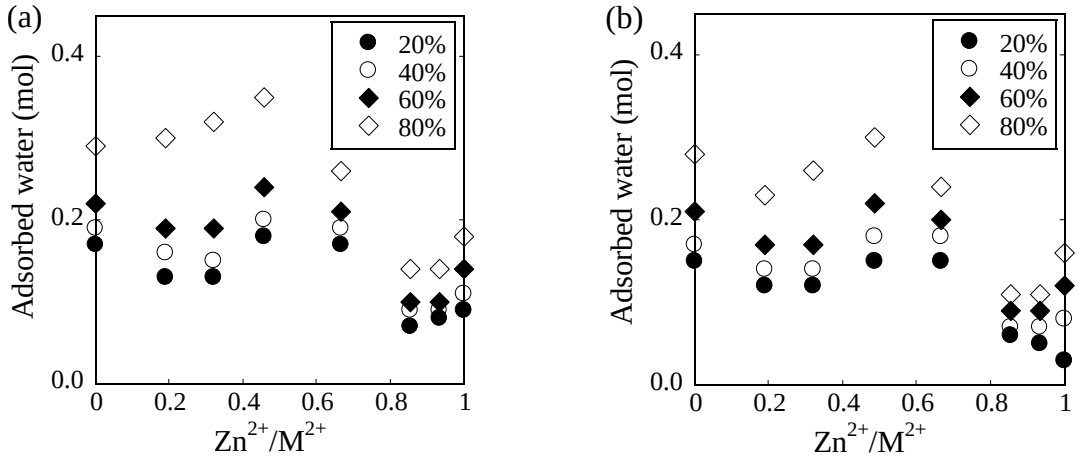
Supplementary information



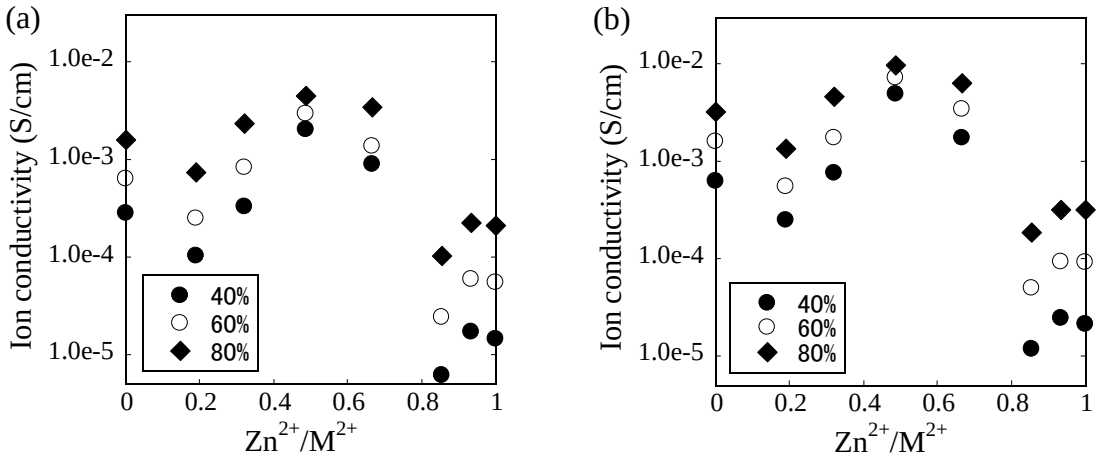
SI Figure 1. TEM images of $(Mg_{(1-x)}Zn_x)_2Al\text{-}CO_3^{2-}$LDH powder with different Zn^{2+}:divalent metals ratios: (a) 0, (b) 0.19, (c) 0.32, (d) 0.49, (e) 0.67, (f) 0.86, (g) 0.94 and (h) 1.00.



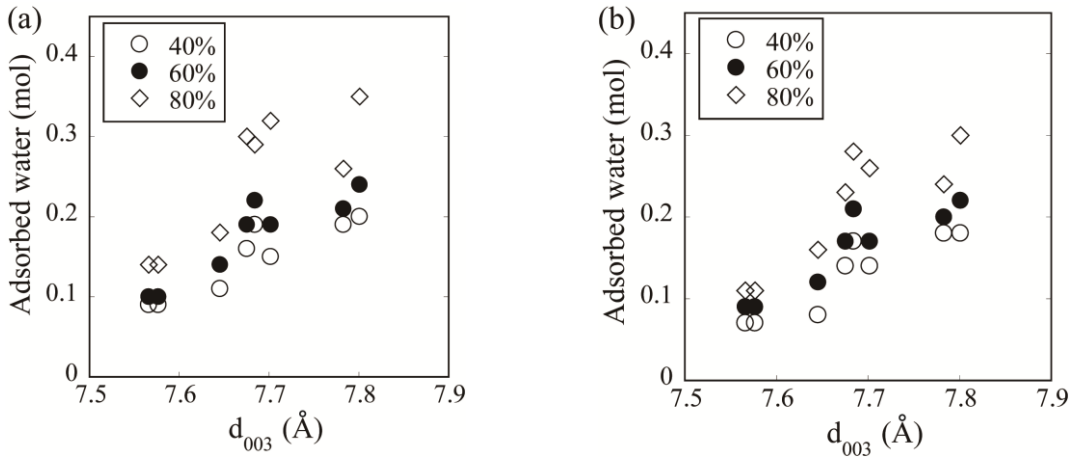
SI Figure 2. Plot of zeta potential of $(Mg_{(1-x)}Zn_x)_2Al\text{-}CO_3^{2-}$LDHs *vs.* the Zn^{2+}:divalent metals ratio.



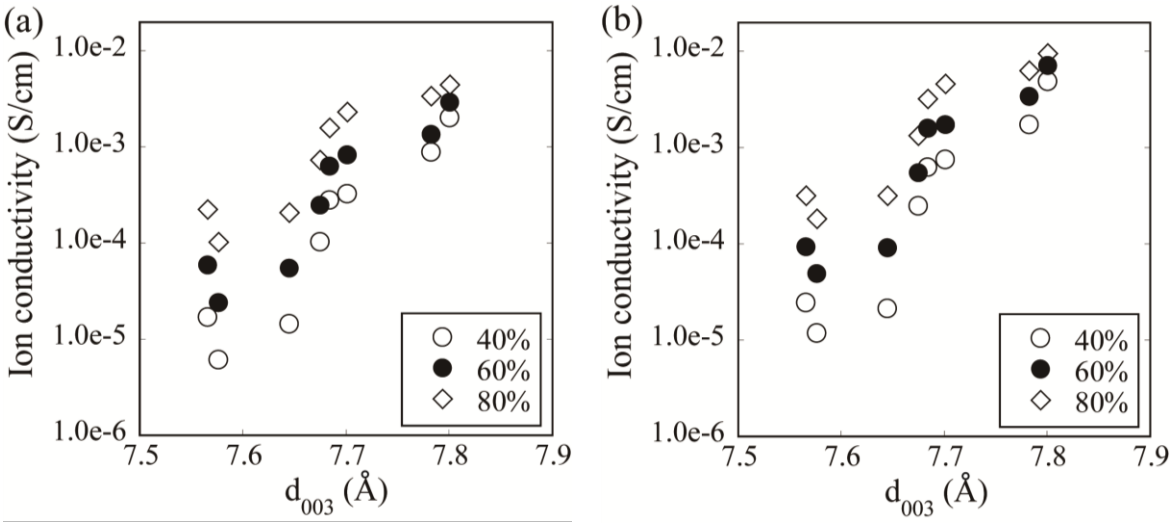
SI Figure 3. Amount of adsorbed water per unit molecule, $[(Mg_{(1-x)}Zn_x)_yAl_z(OH)_2](CO_3^{2-})_{z/2}\cdot nH_2O$ under various humidity conditions at (a) 50 °C and (b) 80 °C, where $y+z=1$.



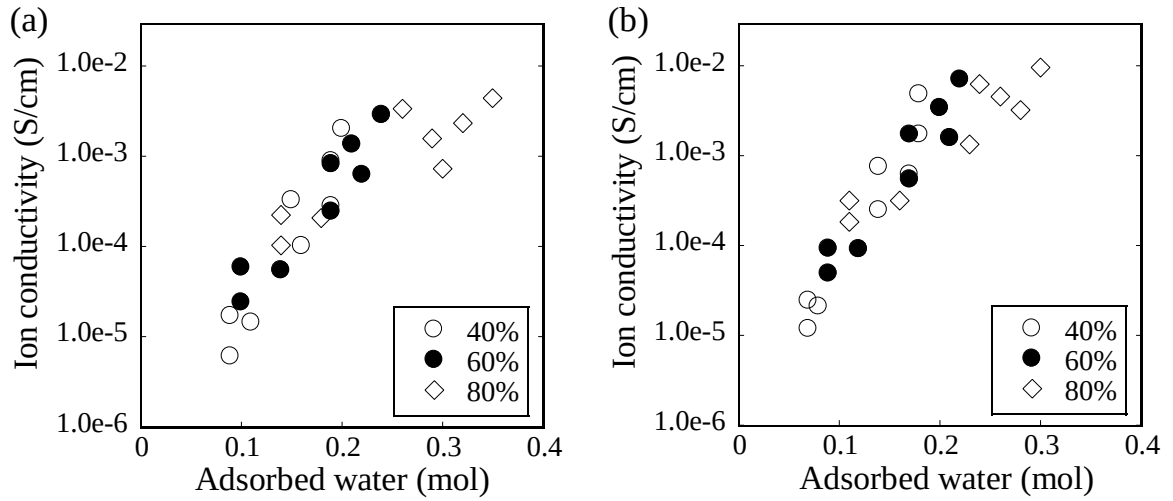
SI Figure 4. Ion conductivity of each LDH with different Zn^{2+}:divalent metals ratio under various humidity conditions at (a) 50 °C and (b) 80 °C.



SI Figure 5. Amount of adsorbed water in unit molecule, $[(Mg_{(1-x)}Zn_x)_yAl_z(OH)_2](CO_3^{2-})_{z/2}\cdot nH_2O$ at different humidities at (a) 50 °C and (b) 80 °C and interlayer distances (d_{003}), where $y+z=1$.



SI Figure 6. Ion conductivity under various humidity conditions at (a) 50 °C and (b) 80 °C and interlayer distance (d_{003}) at room temperature.



SI Figure 7. Ion conductivity and adsorbed water under different humidities at (a) 50 °C and (b) 80 °C

SI Table 1. Activation energy of ion conduction in $(Mg_{(1-x)}Zn_x)_2Al\text{-}CO_3^{2-}$LDHs under R.H 80 % from 30 °C to 80 °C, calculated from the Arrhenius plot based on the reference S-1.

Zn^{2+}:divalent metals ratio (x)	Activation energy (kJ/mol)	Zn^{2+}:divalent metals ratio (x)	Activation energy (kJ/mol)
0	25	0.67	30
0.19	23	0.86	22
0.32	23	0.94	27
0.49	26	1.00	20

References

S-1. N. Hara, H. Ohashi, T. Ito and T. Yamaguchi, *J Phys Chem B*, 2009, **113**, 4656-4663.