

Supporting information

Table S1. Chemical formulae of MgAl-LDH treated with various concentration of CoCl₂ solution under room temperature for 24 h (Metal ratio was obtained from ICP-AES measurement)

[CoCl ₂ (aq)]	Chemical formula
0.02 M	Mg _{2.56} Co _{0.01} Al(OH) _{7.14} (CO ₃) _{0.5} ·mH ₂ O
0.4 M	Mg _{2.51} Co _{0.02} Al(OH) _{7.06} (CO ₃) _{0.5} ·mH ₂ O
0.8 M	Mg _{2.51} Co _{0.04} Al(OH) _{7.1} (CO ₃) _{0.5} ·mH ₂ O

At room temperature, the Co²⁺ substitution for Mg²⁺ in MgAl-LDH lattice was not significant during 24 h. Only a slight increase in cobalt content was observed with respect to concentration of Co²⁺. However, the cobalt content in LDH lattice was negligible. According to the Musumeci *et al.*'s report,¹ LDHs treated with aqueous divalent ions showed substitution at very long reaction time ~ 5 days. Short reaction time of 24 h was thought to be not enough for the effective substitution. However, we found that Co²⁺ substitution was considerable within 3 h at hydrothermal condition. This result implied that hydrothermal condition was essential for the isomorphous substitution reaction between solid LDH and aqueous divalent ion.

Table S2. Peak positions of (110) and lattice parameter *a* values for pristine LDH and Co-substituted LDHs

Sample name	Position of (110) (2 θ)	Lattice parameter a (Å)
Pristine LDH	60.71	3.046
LDH-Co-1	60.76	3.043
LDH-Co-3	60.91	3.033
LDH-Co-6	60.81	3.053
LDH-Co-12	60.35	3.036
LDH-Co-24	60.42	3.053

The (110) peak positions and lattice parameter a values slightly increased during 3 hours and decreased afterwards within a small range ~ 0.01 Å. According to Tok *et al.*'s report,² the lattice parameter slightly decreased for first 6 h and increased afterwards during hydrothermal treatment of cerium oxide nanoparticles. This was explained by the formation of stable crystal during hydrothermal treatment. Our results was similar to those of Tok *et al.*'s, and thus the slight change in lattice parameter in Co-substituted LDH was attributed to the stabilization of crystal during hydrothermal treatment.

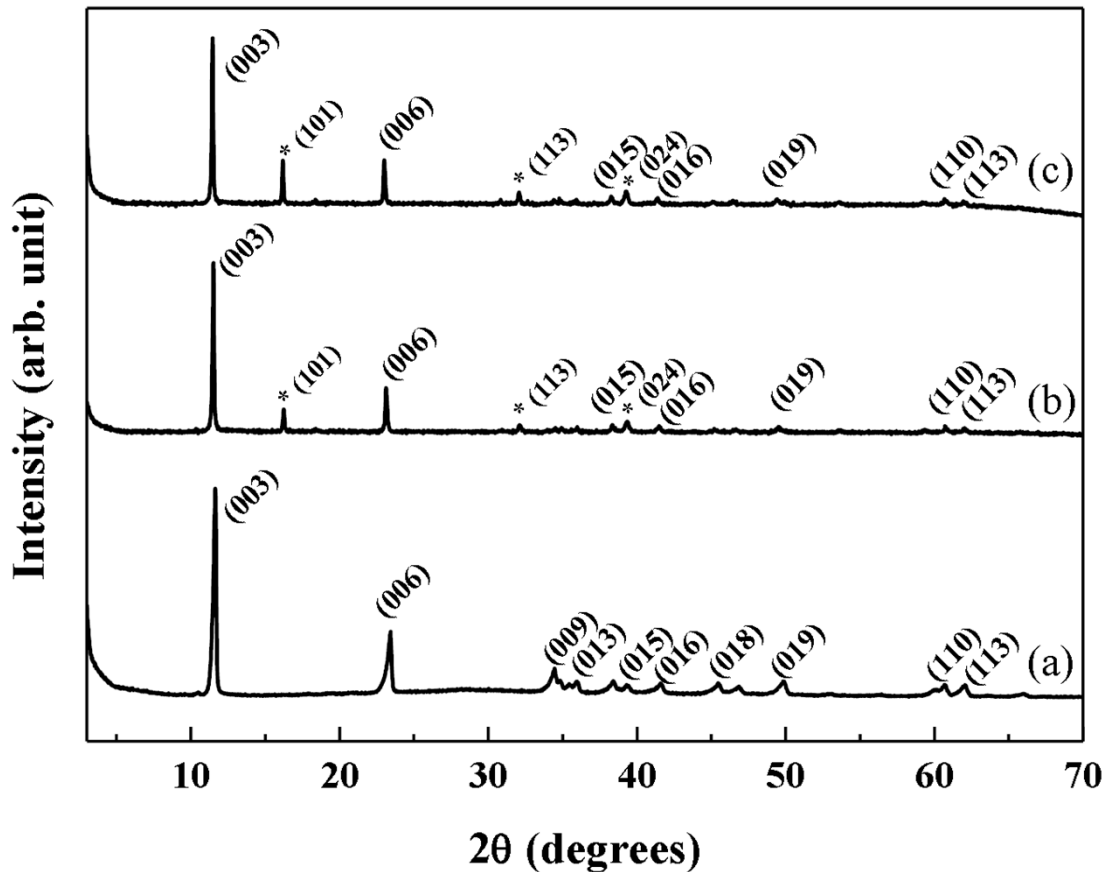


Fig. S1 Powder X-ray diffraction patterns of (a) pristine LDH and hydrothermally treated pristine LDH with (b) 0.4 M Co^{2+} solution and (c) 0.8 M Co^{2+} solution during 24 hours (* indicated cobalt hydroxychloride ($\text{Co}_2(\text{OH})_3\text{Cl}$) phase (JCPDS No. 73-2134))

The XRD patterns of MgAl-LDHs, which were treated with high concentration of CoCl_2 solutions (0.8 and 0.4 M) under hydrothermal conditions at 150 °C for 24 h, showed evolution of impurity phases. According to the JCPDS database, newly emerged peaks at 16.2, 32.1 and 39.3 ° were identified as (101), (113) and (024) reflections of cobalt hydroxychloride ($\text{Co}_2(\text{OH})_3\text{Cl}$, JCPDS No. 73-2134). On the other hand, there was no impurity peaks detected with LDHs treated with 0.02 M CoCl_2 solution. In high Co^{2+} solution, the surface precipitation of Co^{2+} through hydrolysis might be prevalent to substitution reaction, resulting in the formation of unintended cobalt impurity phase.

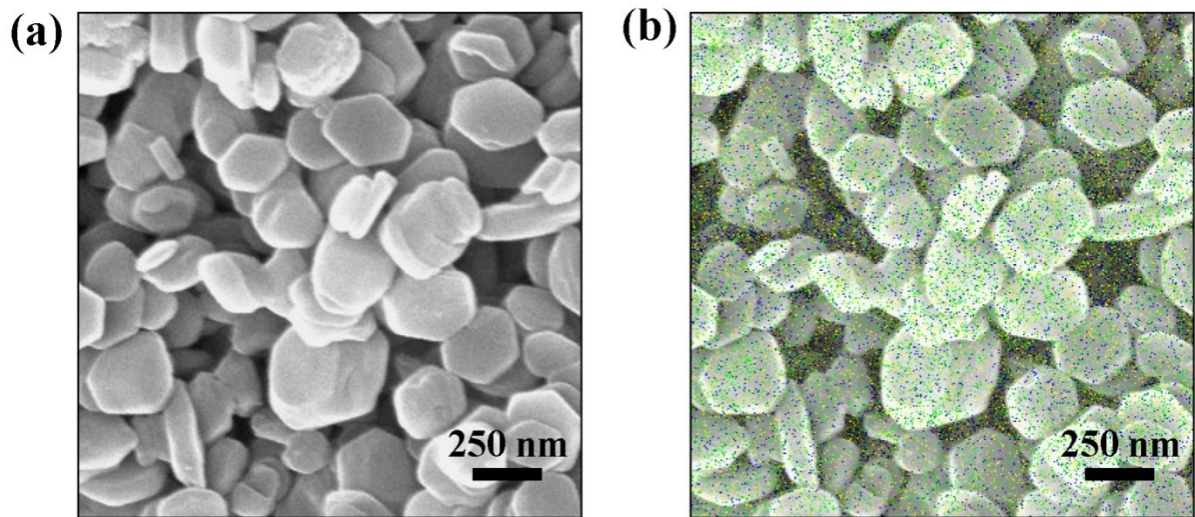


Fig. S2. (a) Scanning electron microscopy (SEM) image and (b) energy dispersive spectroscopy (EDS) mapping image of LDH-Co₂₄ (blue: Mg, yellow: Al, green: Co)

From the SEM-EDS mapping results, we confirmed that the cobalt ions were homogeneously distributed all over the particles. This stood for that the Co²⁺ ions evenly substituted for Mg²⁺ ions, not showing surface precipitates nor localized substitution.

Reference

1. A. W. Musumeci, T. L. Schiller, Z. P. Xu, R. F. Minchin, D. J. Martin and S. V. Smith, *J. Phys. Chem. C*, 2009, **114**, 734-740
2. A.I.Y. Tok, F.Y.C. Boey, Z. Dong and X.L. Sun, *J. Mater. Process. Technol.*, 2007, **190**, 217-222