Supplementary Information

Tuning the Structural Properties of Cadmium-Aluminum Layered Double Hydroxide for Enhanced Photocatalytic Dye Degradation

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Figure S1. Energy dispersive X-ray (EDX) spectra of CdAl LDH having a cationic ratio of x = 0.25 (A), x = 0.28 (B) and x = 0.33 (C).



Figure S2. Transmission electron microscopy (TEM) image of CdAl LDH with a cationic ratio of 0.25 at different magnifications (A, B) and the corresponding selected area electron diffraction (SAED) pattern that reveals two faint rings (dashed red lines) corresponding to the (003) and (006) inter-layer periodicity of LDH.



Figure S3. Thermal gravimetric analysis spectra of CdAl LDH having a cationic ratio of x = 0.33 (i), x = 0.28 (ii) and x = 0.25 (iii).



Figure S4. ²⁷Al solid-state NMR of CdAl LDH having a cationic ratio of x = 0.33 (A), x = 0.28 (B) and x = 0.25 (C).



Figure S5. Nitrogen adsorption (solid circle)-desorption (empty circle) isotherms at 77 K of CdAl LDHs having x = 0.33 (A), 0.28 (B) and 0.25 (C).



Figure S6. Survey XPS spectra of the CdAl-LDH having having x = 0.25 (A), 0.28 (B) and 0.33 (C). XPS spectra of Cd 3d (D), with its corresponding Auger electron analysis (E), Al 2p (F) and O 1s (G) peaks of CdAl LDH having x = 0.25. XPS spectra of Cd 3d (H), with its corresponding Auger electron analysis (I), Al 2p (J) and O 1s (K) peaks of CdAl LDH having x = 0.28. XPS spectra of Cd 3d (L), with its corresponding Auger electron analysis (M), Al 2p (N) and O 1s (O) peaks of CdAl LDH having x = 0.33.



Figure S7. UV-Vis diffuse reflectance spectra converted into $(\alpha h\nu)^2$. E_g is obtained by extrapolating the linear region straight line (dashed line) to the *h*v axis intercept of the $(\alpha h\nu)^2$ vs. *h*v plot. The band gap values for CdAl LDH with a cationic ratio of x = 0.33 (A), x = 0.28 (B) and x = 0.25 (C) are calculated to be E_g = 2.61 ev, 2.53 ev and 2.35 ev, respectively.



Figure S8. UV-vis absorption spectra of the MB solution during its decomposition reaction under visible light (A) and in the presence of CdAl LDH having a cationic ratio of x = 0.33 (B), x = 0.28 (C) and x = 0.25 (D).



Figure S9. UV-vis absorption spectra of the MB solution in the absence of light and in the presence of CdAl LDH having a cationic ratio of x = 0.33 (A), x = 0.28 (B) and x = 0.25 (C). The MB adsorption reaches its maximum at 20 min for both x = 0.33 and x = 0.28 while for x = 0.25 the saturation takes around 40 min (D).



Figure S10. UV-vis absorption spectra of the MB solution during its decomposition reaction under visible light in the presence of $Cd(OH)_2$ (A) and $Al(OH)_3$ (B). Photocatalytic performance of $Cd(OH)_2$ and $Al(OH)_3$ in the degradation of MB showing minimal efficiency (C).



Figure S11. PXRD patterns of CdAl LDHs before photodegradation (x = 0.33 (A(i)), x = 0.28 (A(iii)) and x = 0.25 (A(v))) and after 4 cycles of photodegradation (x = 0.33 (A(ii)), x = 0.28 (A(iv)) and x = 0.25 (A(vi))). SEM images of the regenerated CdAl LDHs, with cationic ratio of x = 0.33 (B), x = 0.28 (C) and x = 0.25 (D), after four cycles of photodegradation.