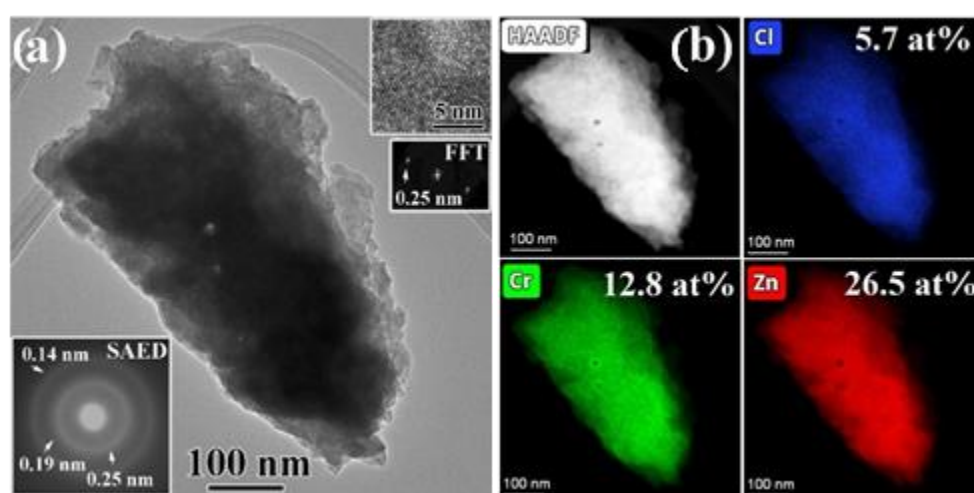
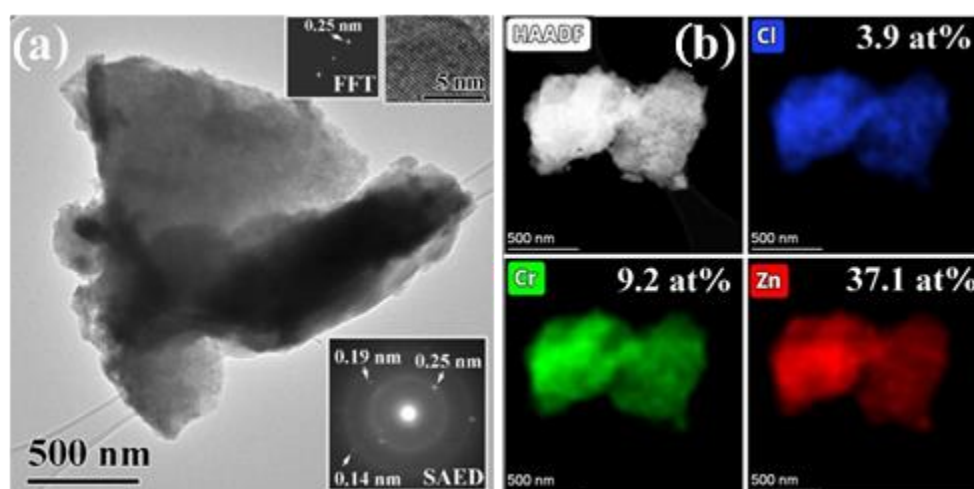


## SUPPORTING INFORMATION

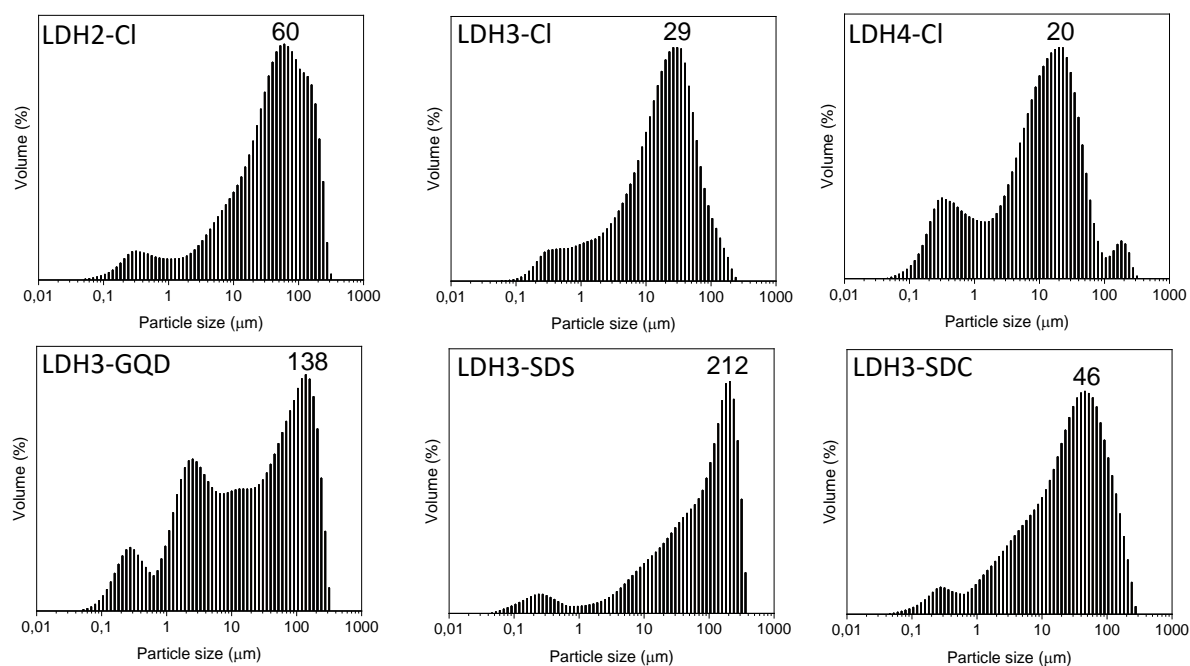
### Zn-Cr layered double hydroxides for photocatalytic transformation of CO<sub>2</sub> under visible light irradiation: The effect of the metal ratio and interlayer anion



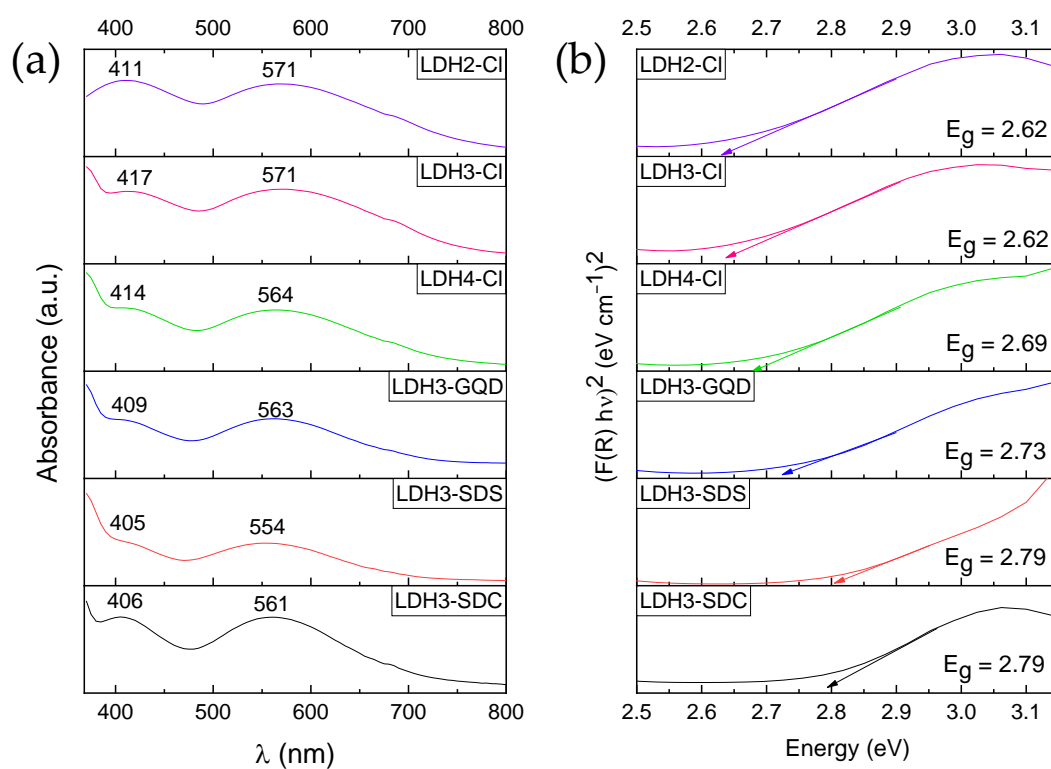
**Figure S1.** (a) TEM images of LDH2-Cl including a SAED pattern and HRTEM imaging (with its corresponding FFT). (b) EDS elemental mapping of LDH2-Cl.



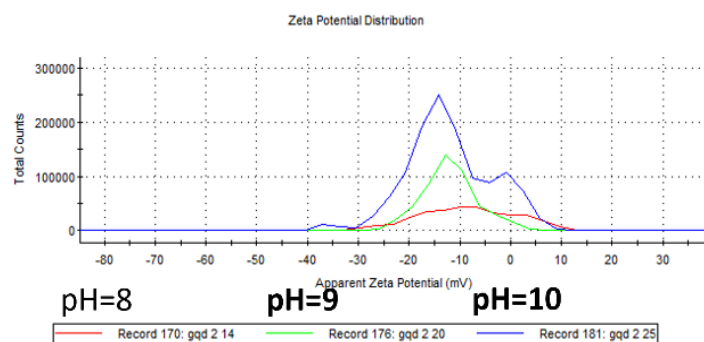
**Figure S2.** (a) TEM images of LDH4-Cl including a SAED pattern and HRTEM imaging (with its corresponding FFT). (b) EDS elemental mapping of LDH4-Cl.



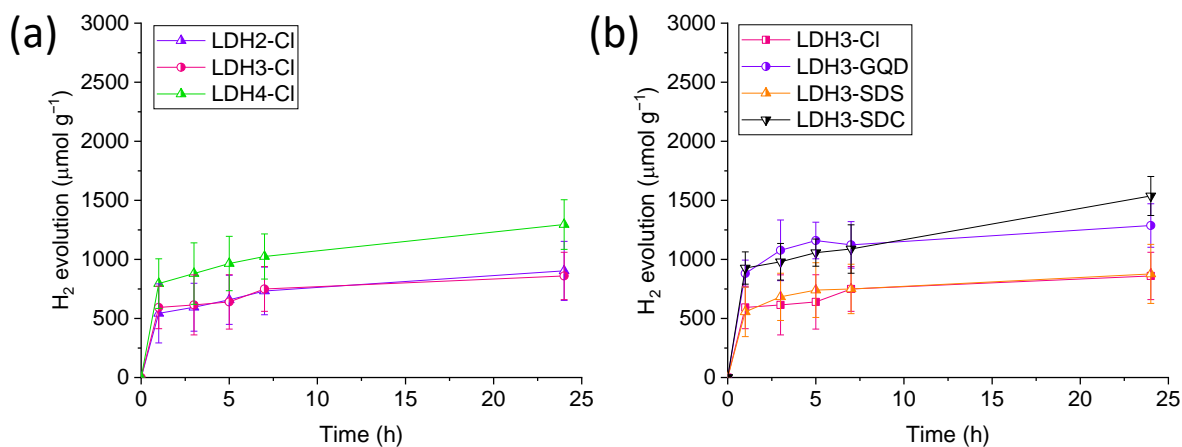
**Figure S3.** Particle size distribution of Zn-Cr LDH based materials determined by laser diffraction.



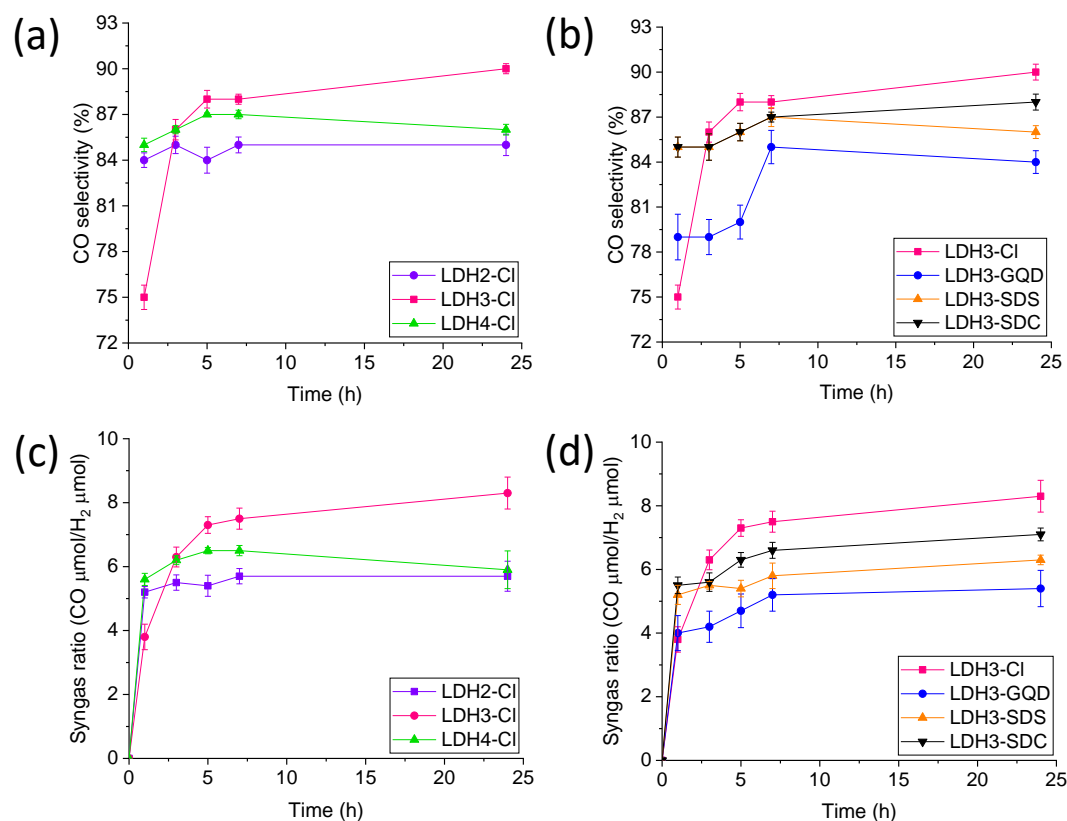
**Figure S4.** (a) UV-vis absorption spectra and (b) band gaps of Zn-Cr LDH based materials.



**Figure S5.** Zeta potential distributions at different pH for GQD.



**Figure S6.** Hydrogen evolution for the photocatalytic systems: (a) using different metal ratio and chloride as interlayer anion and (b) using different interlayer anion and metal ratio of ca. 3.



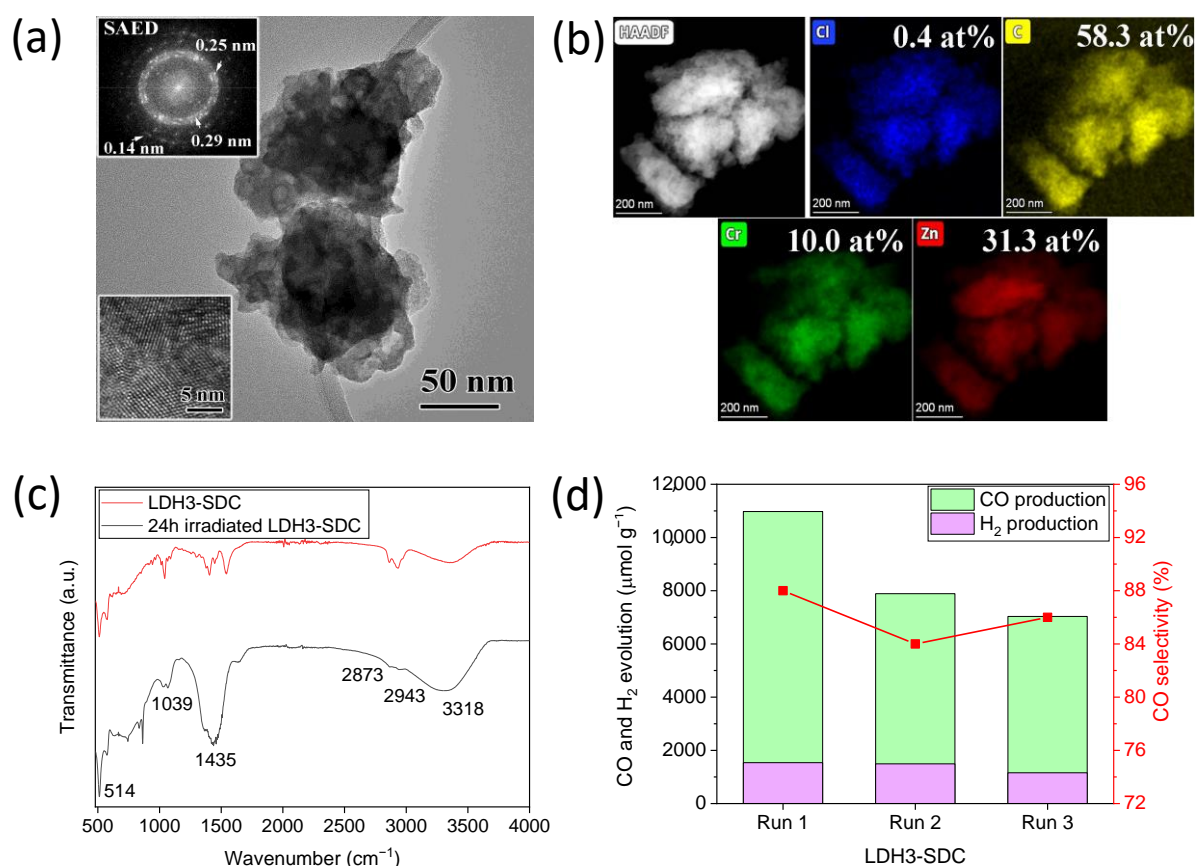
**Figure S7.** CO selectivity and syngas ratio with error bars for the photocatalytic systems: (a and c) using different metal ratio and chloride as interlayer anion and (b and d) using different interlayer anion and metal ratio of ca. 3.

**Table S1.** Apparent quantum yield (CO) for all photocatalytic systems using Zn-Cr LDH based materials as catalysts irradiated at 450 nm for 24 h.

Material	AQY (%)
LDH2-Cl	1.1
LDH3-Cl	1.6
LDH4-Cl	1.7
LDH3-GQD	1.6
LDH3-SDS	1.2
LDH3-SDC	2.4

**Table S2.** Photocatalytic performance of different LDH materials as catalyst in visible light CO<sub>2</sub> photoreduction systems with Ru(bpy)<sub>3</sub><sup>2+</sup> as photosensitizer and TEOA as sacrificial electron donor.

Material	Light source	Time reaction (h)	CO production (mmol g <sup>-1</sup> )	CO selectivity (%)	Reference
LDH3-SDC	Penn PhD Photoreactor M2 λ=450 nm	1	5.03	88	This work
CoAl-LDH	300 W xenon lamp (λ > 400 nm)	1	2.52	61	[33]
NiAl-LDH	300 W xenon lamp (λ > 400 nm)	1	0.26	82	[33]
ZnAl-LDH	300 W xenon lamp (λ > 400 nm)	1	0.60	57	[33]
MgAl-LDH	300 W xenon lamp (λ > 400 nm)	1	0.70	29	[33]
CoMgAl-LDH	300 W xenon lamp (λ > 400 nm)	1	7.70	57	[34]
NiAl-LDH	300 W xenon lamp (λ > 400 nm)	1	0.20	80	[35]

**Figure S8.** (a) TEM image including a SAED pattern and HRTEM imaging, (b) EDS elemental mapping and (c) FTIR-ATR spectra of LDH3-SDC after 24 h under visible-light irradiation. (d) CO and H<sub>2</sub> evolution and CO selectivity in 3 runs.

## References

33. Bai, S.; Wang, Z.; Tan, L.; Waterhouse, G.I.N.; Zhao, Y.; Song, Y.-F. 600 Nm Irradiation-Induced Efficient Photocatalytic CO<sub>2</sub> Reduction by Ultrathin Layered Double Hydroxide Nanosheets. *Ind. Eng. Chem. Res.* **2020**, *59*, 5848–5857, doi:10.1021/acs.iecr.0c00522
34. Ning, C.; Wang, Z.; Bai, S.; Tan, L.; Dong, H.; Xu, Y.; Hao, X.; Shen, T.; Zhao, J.; Zhao, P.; et al. 650 Nm-Driven Syngas Evolution from Photocatalytic CO<sub>2</sub> Reduction over Co-Containing Ternary Layered Double Hydroxide Nanosheets. *Chem. Eng. J.* **2021**, *412*, 128362, doi:10.1016/j.cej.2020.128362.
35. Wang, Z.; Xu, S.-M.; Tan, L.; Liu, G.; Shen, T.; Yu, C.; Wang, H.; Tao, Y.; Cao, X.; Zhao, Y.; et al. 600 Nm-Driven Photoreduction of CO<sub>2</sub> through the Topological Transformation of Layered Double Hydroxides Nanosheets. *Appl. Catal. B Environ.* **2020**, *270*, 118884, doi:10.1016/j.apcatb.2020.118884.