

Supporting Information

for

Playing with covalent triazine framework tiles for improved CO₂ adsorption properties and catalytic performance

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Additional experimental data



Figure S1: Powder X-ray diffraction analyses of **CTF1–5**, showing their amorphous nature.

Table S1: CHN Elemental Analyses of CTF1-5
samples. All values are given as an average value
over three independent runs.

Sample	N wt %	C wt %	H wt %
CTF1	7.5	65.2	2.2
CTF2	3.6	77.9	1.2
CTF3	29.1	40.0	2.6
CTF4	18.1	50.3	2.4
CTF5	11.4	61.8	2.0





Figure S2: Nitrogen adsorption–desorption isotherms of **CTF1–5** (**A–E**) recorded at 77 K and related pore size distribution evaluated through a NLDFT pore modelling (**A'–E'**). Adsorption-desorption branches of the isotherm recorded for **CTF3** (C) do not close due to incomplete desorption. This effect is likely attributed to a strong material adsorption of N₂ within its very narrow slit-like pores.





Figure S3: XPS N 1s (A–E) and O 1s (A'–E') core level regions of CTF1–5 along with their relative curves fittings



Figure S4: XPS survey spectra recorded for all CTF samples. Zn residues (traces) from the ionothermal synthesis not completely removed from the carefully purified samples during the material work-up are present in **CTF3–5**. The higher the micropore percentage of the CTF sample the higher is the level of Zn contamination (**CTF3** > **CTF4** > **CTF5**). It is important to note that the role of Zn residues with respect to the material performance in EB DDH has already been ruled out in one of our recent publication in the field [1]. Moreover, in mixed oxide-based catalysts derived from hydrotalcites, the higher the Zn content in the catalyst the lower the catalytic performance in DDH process [2]. Overall, Zn residues simply play as innocent spectators with respect to the DDH process.

The % content of all elements was calculated using the corresponding core level peaks properly normalized to the photoemission cross section and assuming a homogeneous distribution arrangement model, and results are given in the table below.

SAMPLE	%C	%O	%Zn	%N	%Na	C/N
CTF1	83.7	8	0	8.3	0	10.1
CTF2	87.9	8.8	0	3.3	0	26.2
CTF3	64.3	10	0.8	24.1	0.8	2.7
CTF4	71.4	10.9	0.3	16.7	0.6	4.3
CTF5	79.1	9.6	0.2	11.1	0.1	7.1

Table of Figure S4: Surface atomic ratios of all elements measured by XPS.



Figure S5: Low-pressure CO₂ adsorption-desorption isotherms for **CTF1–5** at **A**) T = 273 K and **B**) T = 298 K. (solid squares = adsorption isotherms; empty round symbols = desorption isotherms).



Figure S6: Heats of adsorption (Q_{st}) for **CTF1–5** measured from the CO₂ isotherms recorded at 273 and 298 K.



Figure S7: Comparison of the N_2 (black) and CO_2 (red) isotherms at 298 K in the 0–0.1 bar interval for the estimation of the CO_2 vs N_2 selectivity of **CTF1–5** samples through the Henry method.



Figure S8: CO₂ evolution as recorded from the temperature programmed oxidation (TPO) analyses on (A) CTF4 and (B) CTF5 before (top curves) and after (bottom curves) their use in DDH.

References

- 1. Tuci, G.; Pilaski, M.; Ba, H.; Rossin, A.; Luconi, L.; Caporali, S.; Pham-Huu, C.; Palkovits, R.; Giambastiani, G., *Adv. Funct. Mater.* **2017**, *27*, 1605672. doi:10.1002/adfm.201605672
- Balasamy, R. J.; Tope, B. B.; Khurshid, A.; Al-Ali, A. A. S.; Atanda, L. A.; Sagata, K.; Asamoto, M.; Yahiro, H.; Nomura, K.; Sano, T.; Takehira, K.; Al-Khattaf, S. S., *Appl. Catal., A* 2011, *398*, 113-122. doi:10.1016/j.apcata.2011.03.023