Supplementary Information

Straightforward preparation of fluorinated covalent triazine frameworks with significantly enhanced carbon dioxide and hydrogen adsorption capacities

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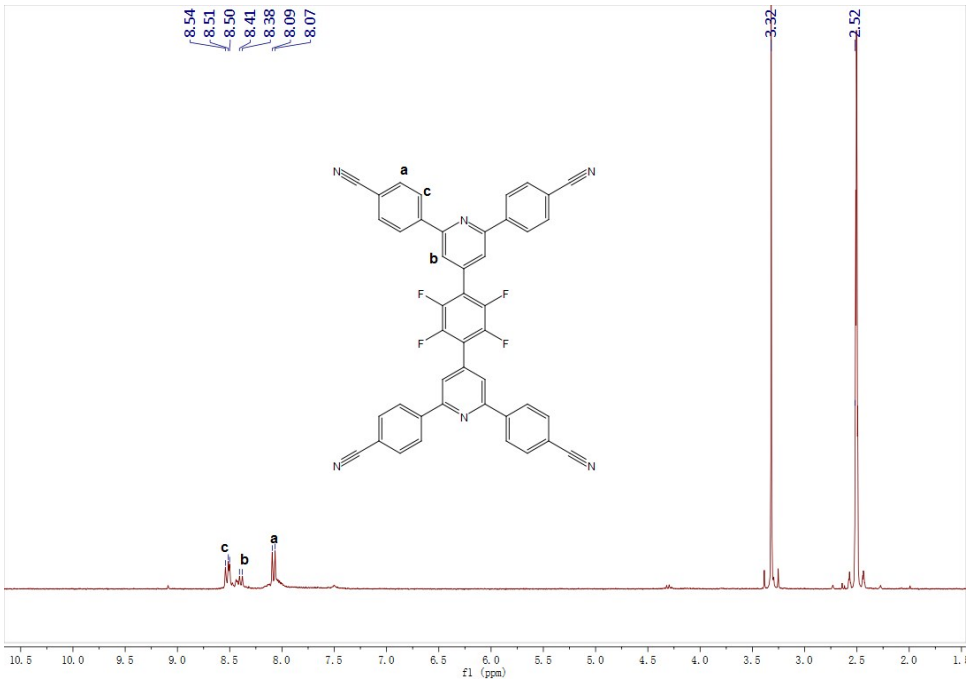

Figure S1. ^{1}H NMR spectrum of the monomer.

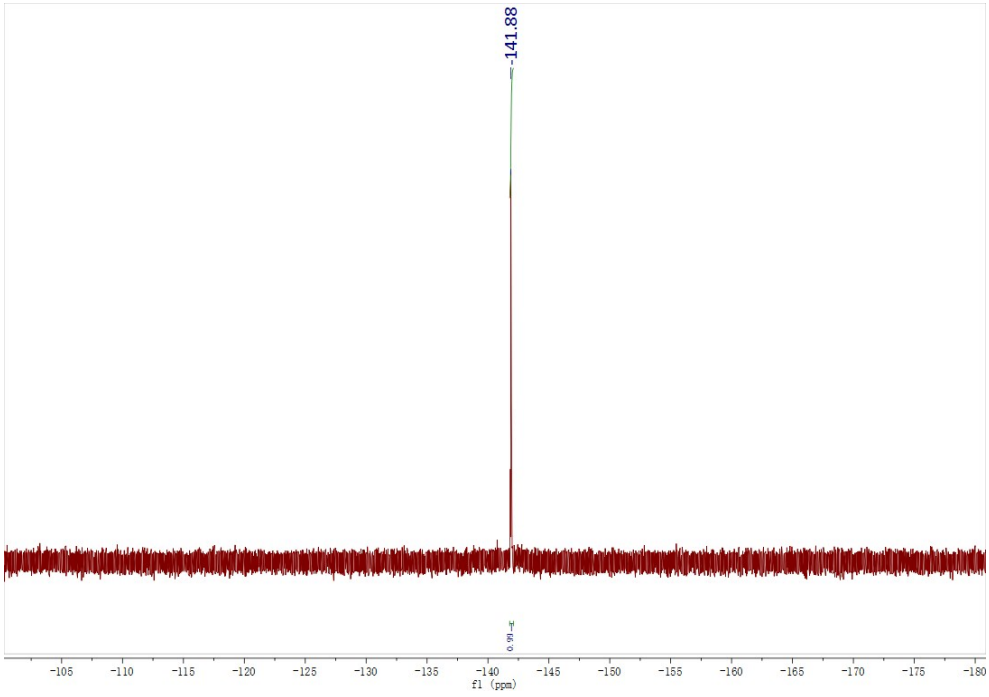

Figure S2. ^{19}F NMR spectrum of the monomer.

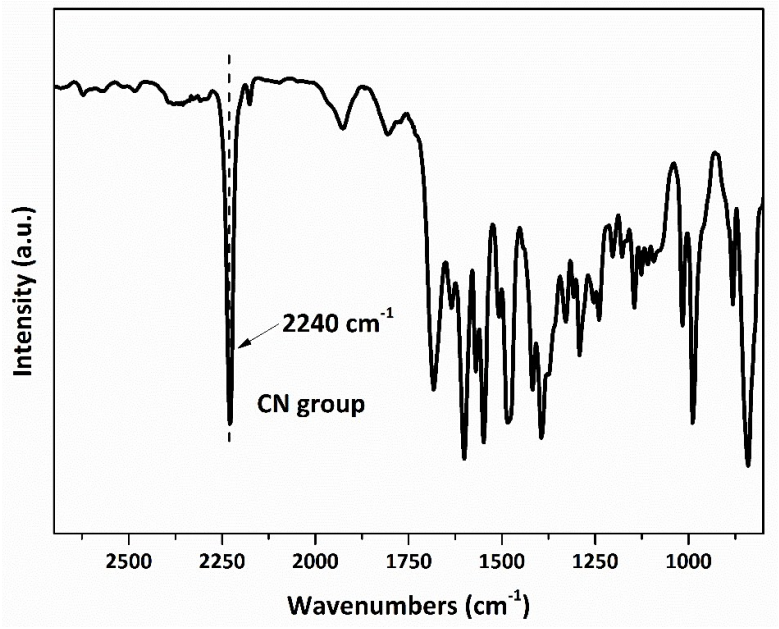

Figure S3. FT-IR spectrum of the monomer.

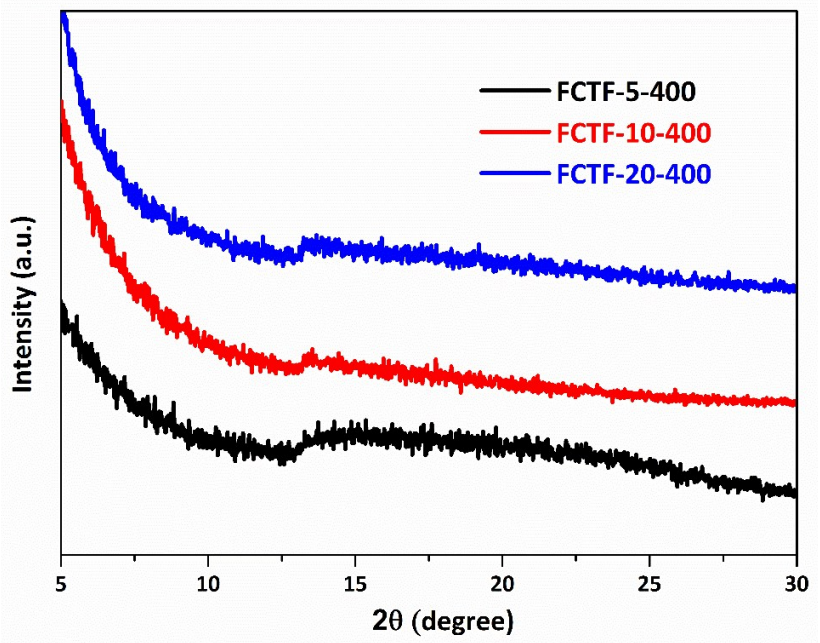

Figure S4. Powder X-ray diffraction (PXRD) patterns of all obtained CTF materials.

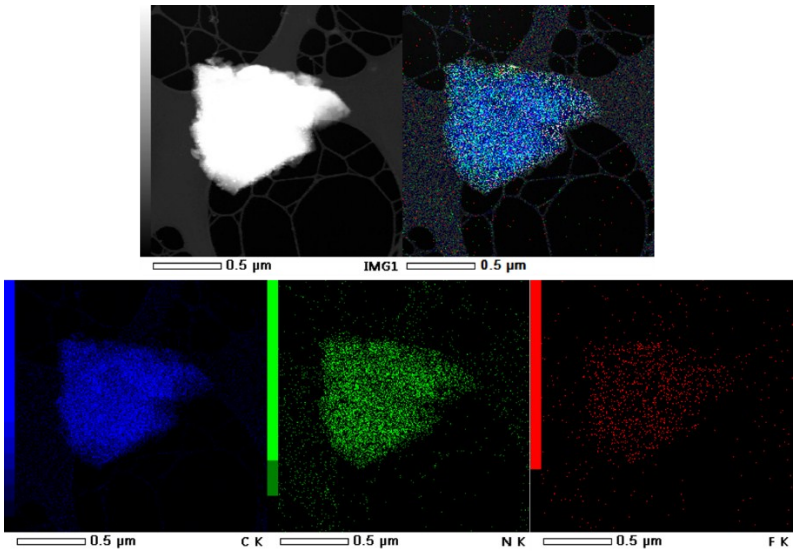

Figure S5. High angle annular dark field scanning-transmission electron microscopy (HAADF-STEM) and corresponding energy dispersive X-ray spectroscopy (EDX) mapping images of carbon (blue), nitrogen (green) and fluorine (red) for the sample FCTF-10-400.

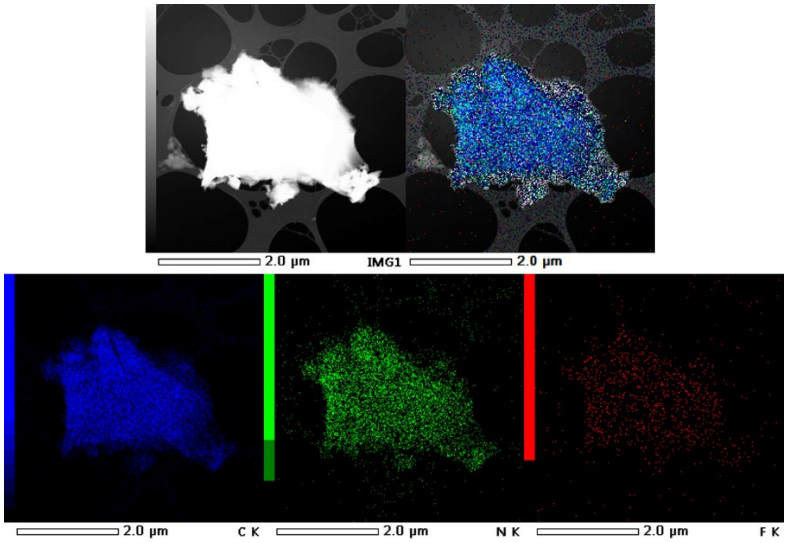

Figure S6. High angle annular dark field scanning-transmission electron microscopy (HAADF-STEM) and corresponding energy dispersive X-ray spectroscopy (EDX) mapping images of carbon (blue), nitrogen (green) and fluorine (red) for the sample FCTF-20-400.

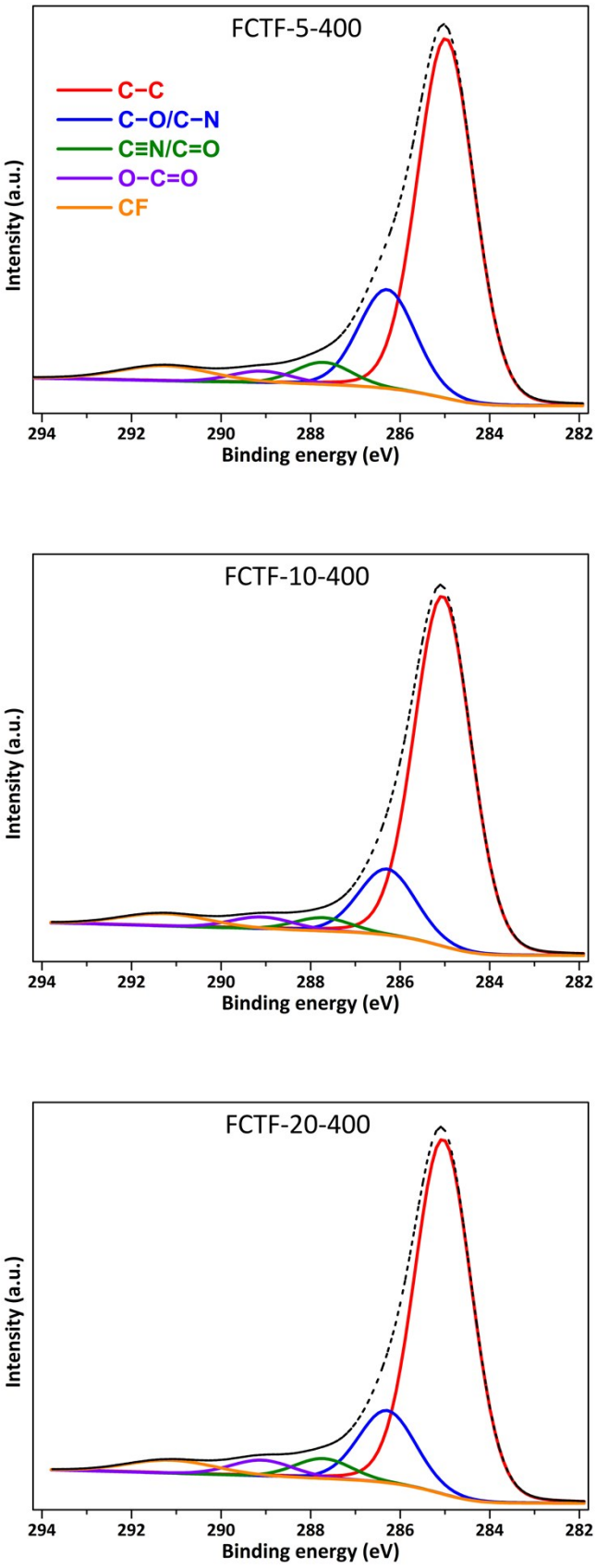

Figure S7. C1s XPS spectra of the fluorinated CTF materials.

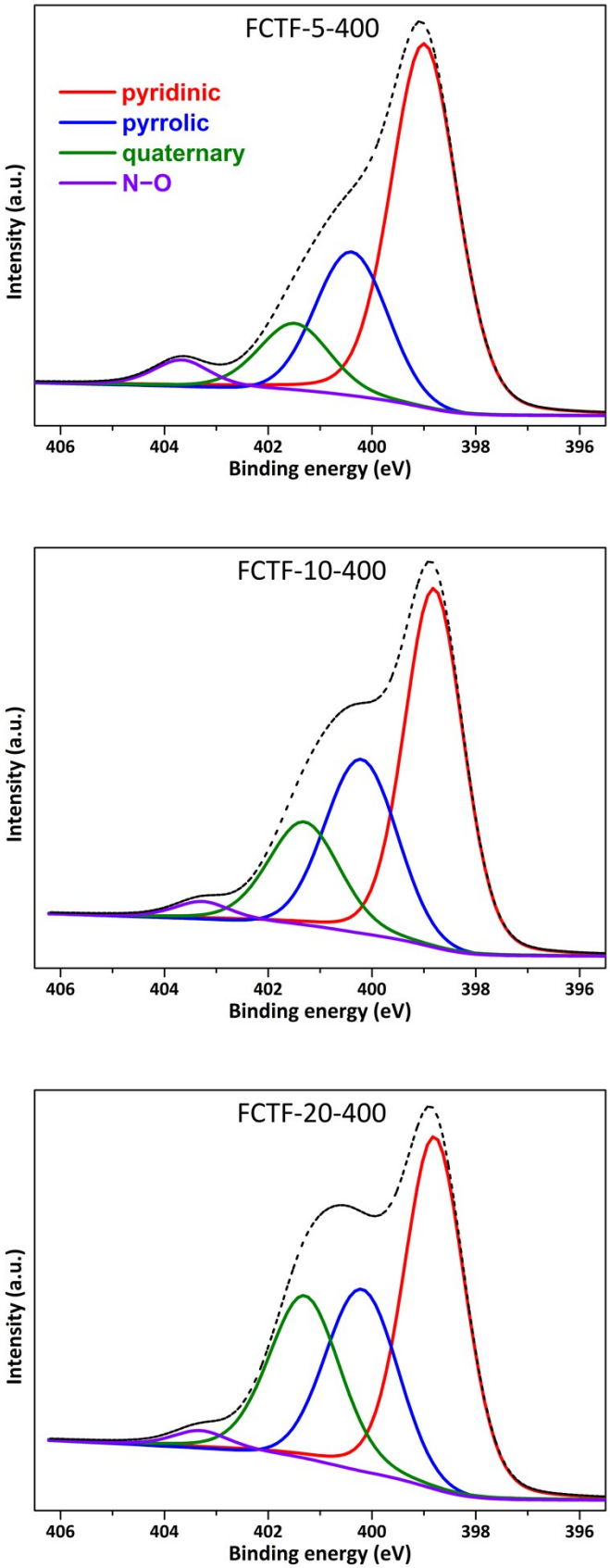

Figure S8. N1s XPS spectra of all studied fluorinated CTFs.

Table S1. Relative concentrations of the different carbon groups present on the fluorinated CTF materials as obtained from C1s curve fitting

Samples	C-C (%)	C-O/C-N (%)	C=-N (%)	O-C=O (%)	CF(%)
FCTF-5-400	69.2	19.0	4.3	2.5	5.0
FCTF-10-400	76.0	14.3	2.9	2.9	4.0
FCTF-20-400	73.4	14.9	4.1	3.6	4.0

Table S2. Relative concentrations of the different nitrogen groups present on the fluorinated CTFs as obtained from N1s curve fitting

Samples	Pyridinic (%)	Pyrrolic (%)	Quaternary (%)	N-O (%)
FCTF-5-400	60.4	23.6	11.9	4.1
FCTF-10-400	51.4	28.0	18.0	2.6
FCTF-20-400	46.6	25.9	25.2	2.3

Table S3. Chemical composition obtained from XPS survey scans for the studied HATN-CTFs

Samples	C (at%)	O (at%)	N (at%)
FCTF-5-400	84.2	7.3	7.1
FCTF-10-400	87.3	4.1	7.7
FCTF-20-400	87.7	5.1	6.7

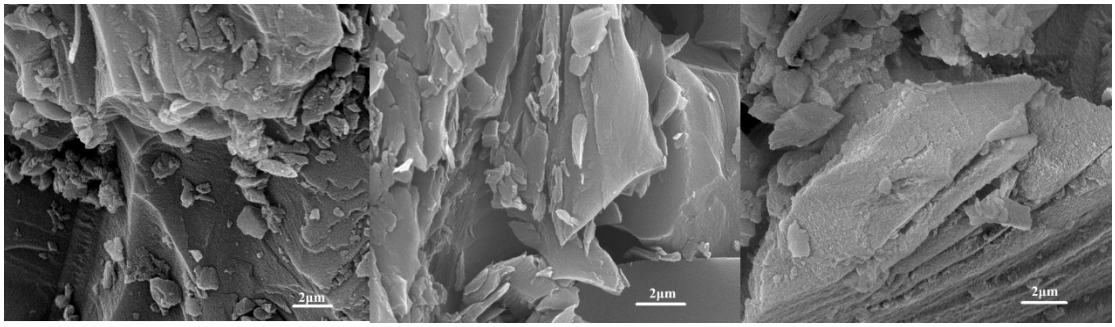

Figure S9. Scanning electron micrographs (SEM) of FCTF-5-400 (left), FCTF-10-400 (middle) and FCTF-20-400 (right).

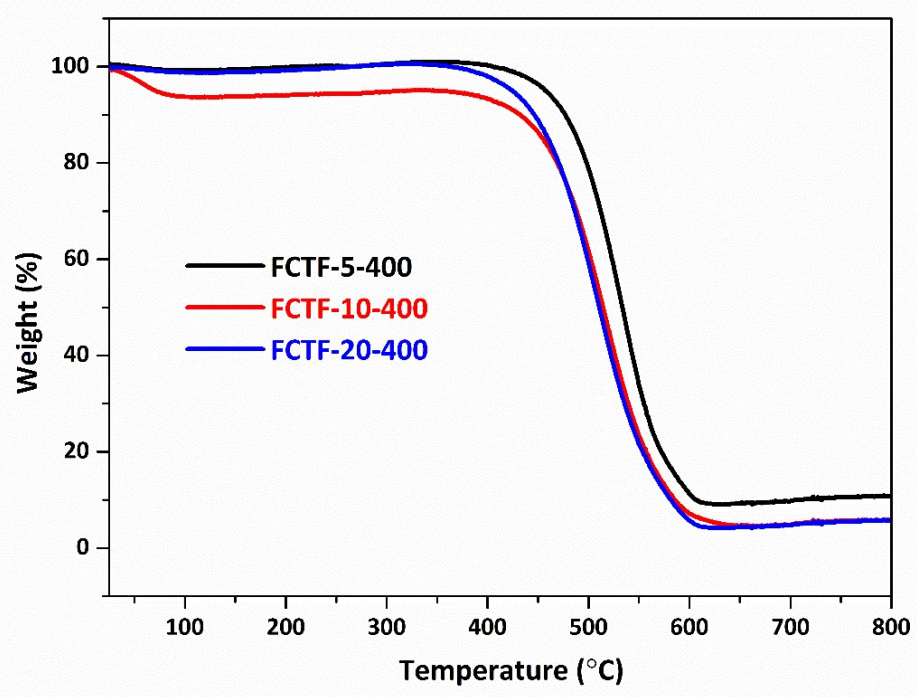

Figure S10. TGA curves of the studied FCTF materials measured at a heating rate of 5 ºC/min under an air flow.

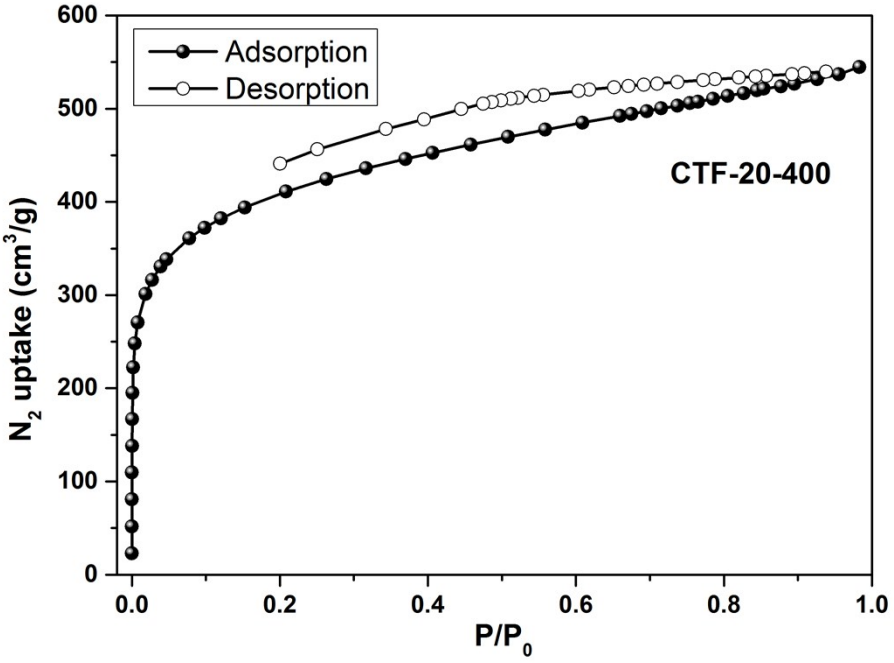

Figure S11. N_2 adsorption (solid symbols) and desorption (empty symbols) isotherm of CTF-20-400.

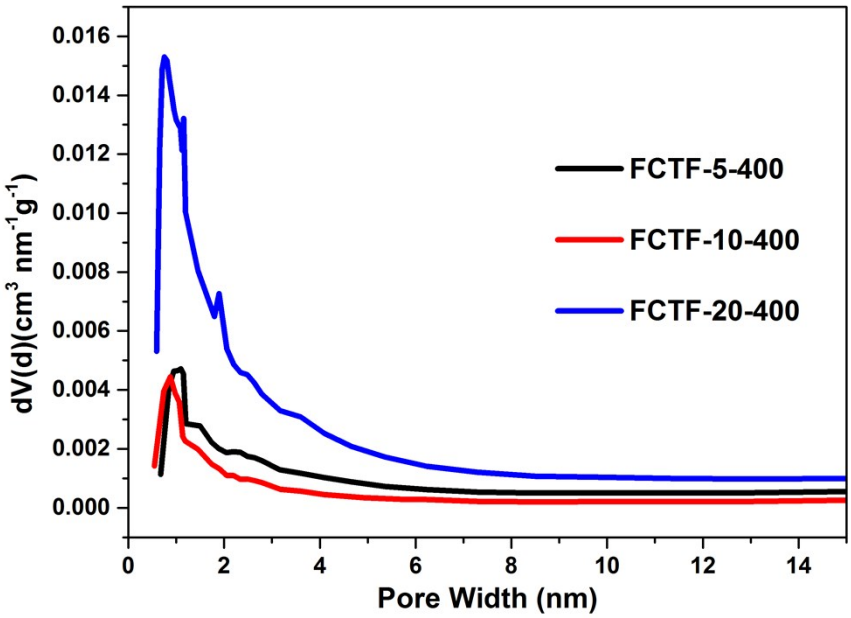

Figure S12. The pore size distributions of the FCTF materials.

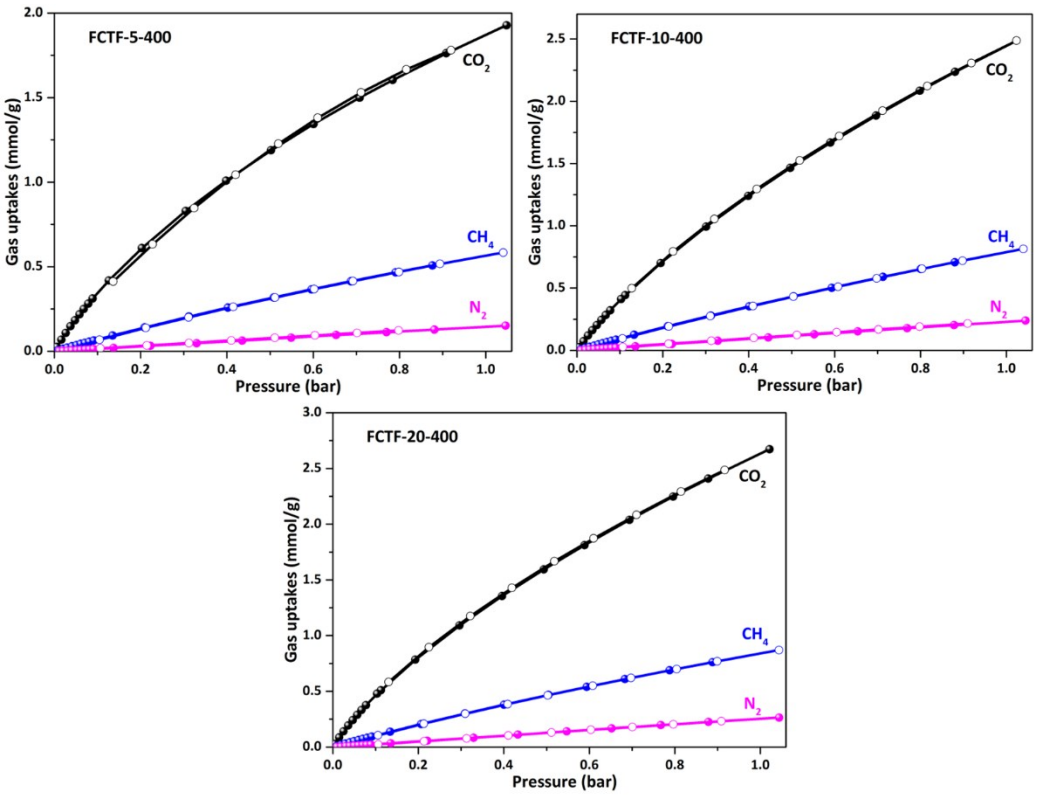

Figure S13. CO_2, CH_4 and N_2 adsorption isotherms for all the obtained fluorinated CTFs measured at 298 K up to 1 bar.

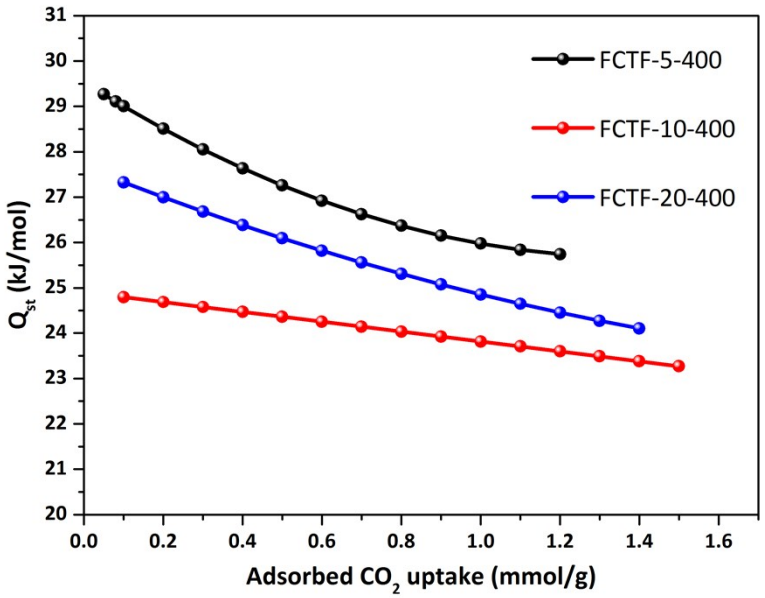

Figure S14. The isosteric heat of CO_2 adsorption (Q_{st}) for all FCTF materials.

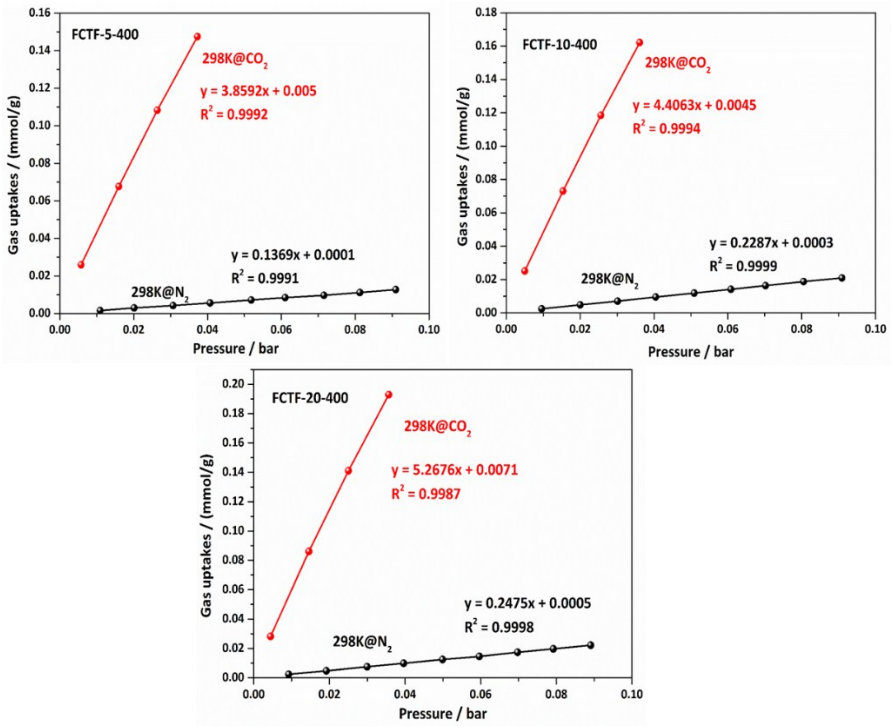

Figure S15. The selectivity of the FCTFs for CO_2 over N_2 isotherms obtained from the initial slope method at 298K.

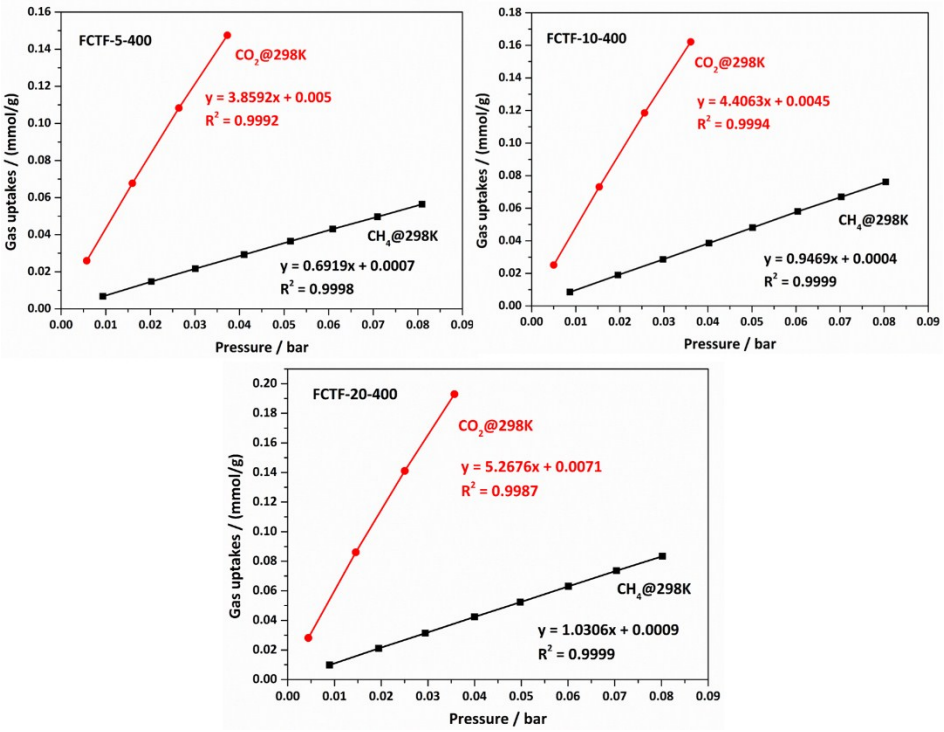

Figure S16. The selectivity of the FCTFs for CO_2 over CH_4 isotherms obtained from the initial slope method at 298K.

FITTING OF ISOTHERMS AND IDEAL ADSORBED SOLUTION THEORY (IAST)

The single component isotherm of CO_2, $CH_4$and N_2, measure at 298 K in the FCTFs were fitted with the single-site Langmuir isotherm model using the equations

$$q = q_{max}\frac{bp}{1+bp}$$

Where, p is the pressure of the bulk gas at equilibrium with the adsorbed phase, q is the adsorbed amount per mass of adsorbent, q_{max} is the maximum capacity of the site and b is affinity coefficient of the site. The adsorption selectivity was calculated using following equations where q_{CO2}, $q_{CH4}$and q_{N2} represent the molar loadings of CTFs that is in equilibrium with a bulk fluid mixture with mole fractions of y_{CO2} = 0.15 and y_{N2} = (1- y_{CO2}) =0.85 for separation of flue gas in ideal conditions (15/85 CO_2/N_2 separation). For CO_2/CH_4 selectivity calculation, y_{CO2} = 0.5 and y_{CH4} = (1- y_{CO2}) =0.5.

$$S = \frac{qCO_2/qN_2}{yCO_2/yN_2}$$

$$S = \frac{qCO_2/qCH_4}{yCO_2/yCH_4}$$

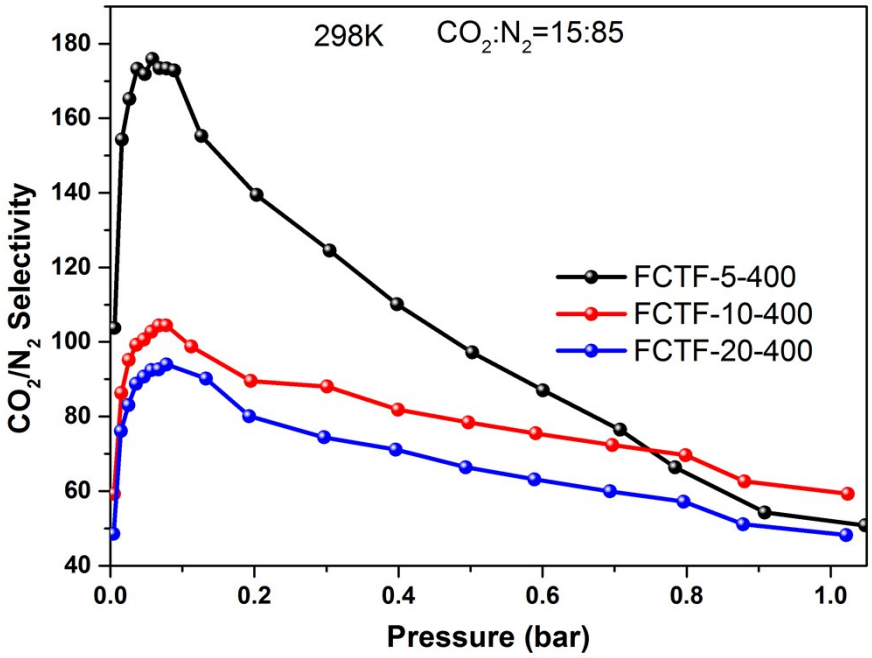

Figure S17. Calculated IAST selectivity of binary gas mixtures (CO_2:N_2 = 15:85) at 298 K for all the studied FCTFs.

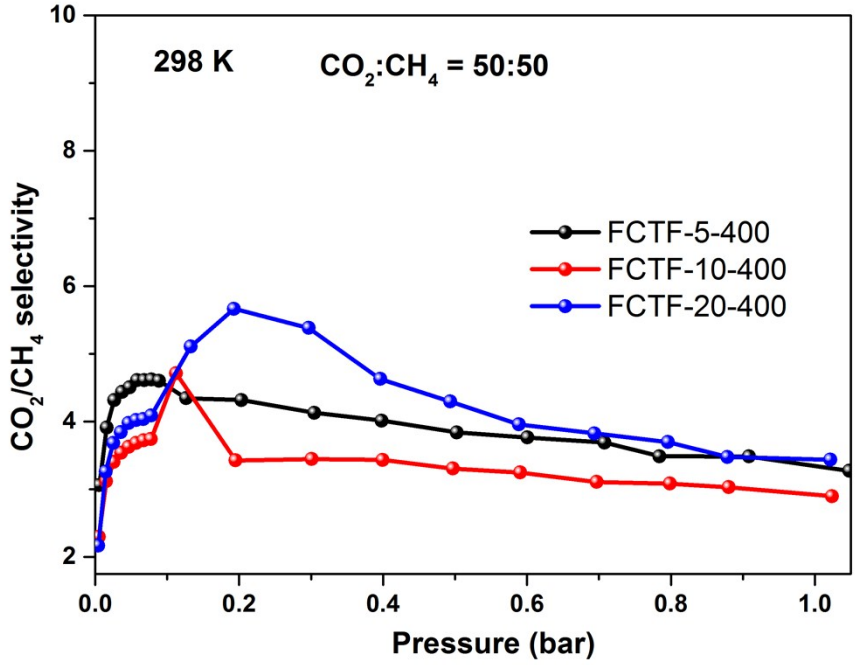

Figure S18: Calculated IAST selectivity of binary gas mixtures (CO_2:CH_4 = 50:50) at 298 K for all the studied FCTFs.