

***Supporting Information***

**Templating of carbon in zeolites under pressure: Synthesis of pelletized zeolite templated carbons with improved porosity and packing density for superior gas (CO<sub>2</sub> and H<sub>2</sub>) uptake properties**

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**Table S1.** Textural properties of zeolite 13X before and after compaction at 740 MPa

Sample	Surface area <sup>a</sup> (m <sup>2</sup> g <sup>-1</sup> )	Pore volume <sup>b</sup> (cm <sup>3</sup> g <sup>-1</sup> )	Pore size <sup>c</sup> (Å)
Z13X	717 (705)	0.33 (0.31)	7.5/10
Z13X@740MPa	692 (680)	0.32 (0.30)	7.5/10

The values in the parenthesis refer to: <sup>a</sup>micropore surface area and <sup>b</sup>micropore volume. <sup>c</sup>pore size distribution maxima obtained from NLDFT analysis.

**Table S2.** CO<sub>2</sub> uptake at 0 °C for zeolite templated carbons templated by powder (CZ13XFAET) or compacted pellets (CZ13XFAETP) of zeolite 13X.

Sample	CO <sub>2</sub> uptake <sup>a</sup> (mmol/g)		Working capacity <sup>b</sup> (mmol/g)
	1 bar	20 bar	
CZ13XFAET	4.3 (83)	23.2 (449)	18.9 (366)
CZ13XFAETP	4.5 (137)	26.0 (789)	21.5 (652)

The values in the parenthesis are volumetric CO<sub>2</sub> uptake in g l<sup>-1</sup>. <sup>a</sup>CO<sub>2</sub> uptake at 25 °C and various pressures (i.e., 1 bar and 20 bar). <sup>b</sup>Defined as the difference of storage capacity between 20 and 1 bar.

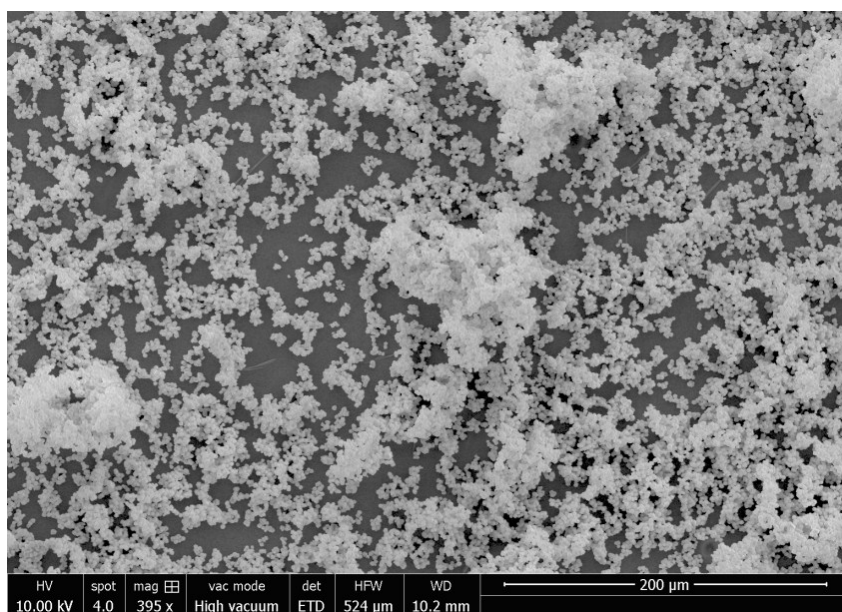
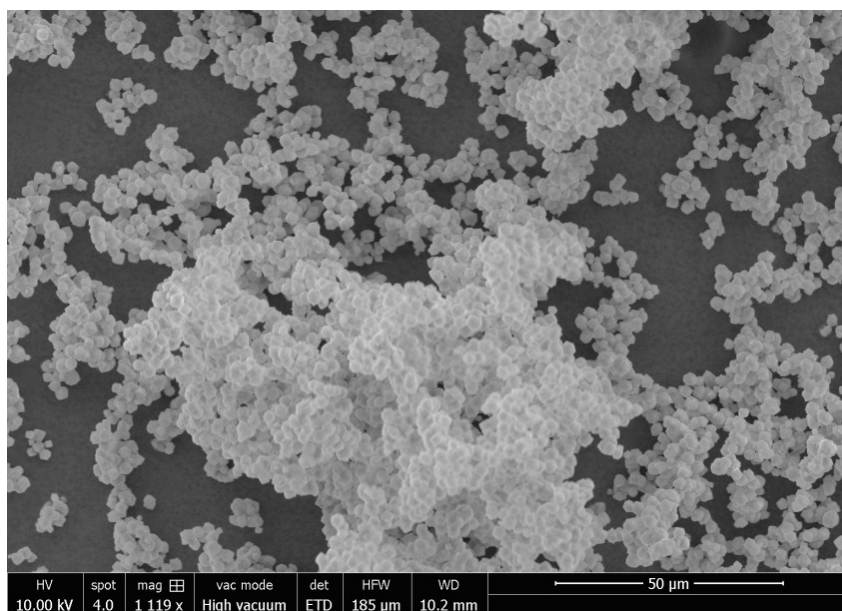
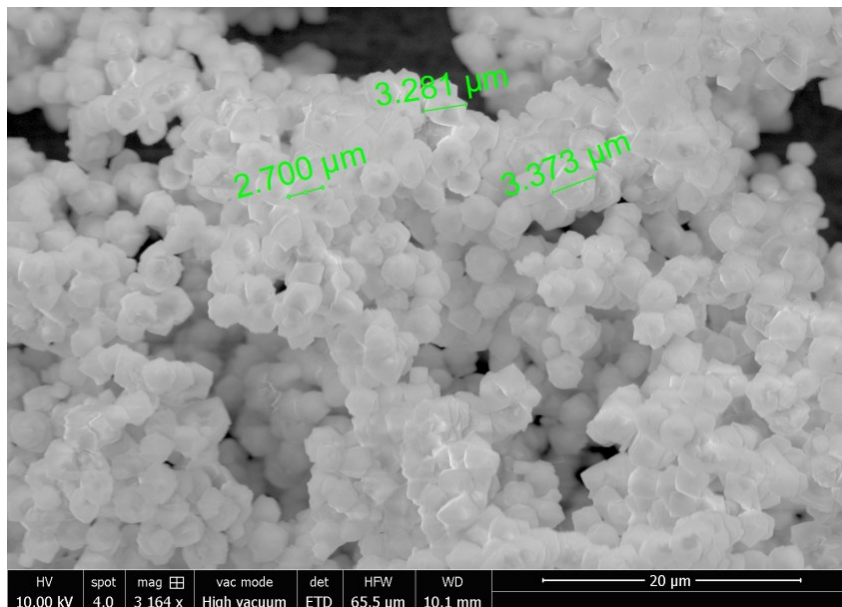
**Table S3.** CO<sub>2</sub> uptake at 25 °C and 20 bar, and working capacity for PSA (20 bar to 1 bar) for powder (CZ13XFAET) and pelletized (CZ13XFAETP) ZTCs compared to top-performing materials reported in the literature.

Material	Packing density (g/cm <sup>3</sup> )	CO <sub>2</sub> uptake (20 bar, 25 °C) (mmol/g)	Working capacity for PSA system <sup>a</sup>			Reference
			(mmol/g)	(g/l)	(cm <sup>3</sup> /cm <sup>3</sup> )	
CZ13XFAETP	0.69	22.0	19.4	589	300	This work
CZ13XFAET	0.44	18.6	16.1	312	159	This work
VR-5 (AC from mesophase pitch)	0.47	26.8	22.0	455	232	1
VR-93 (AC from mesophase pitch)	0.47	30.4	25.6	529	269	1
VR5-4:1 (AC from mesophase pitch)	0.47	22.0	19.4	401	204	2
Maxsorb (comercial AC)	0.29	19.0	16.9	216	110	3
HPC5b2-1100 (hierarchical carbon)		20.8	17.1			4
MOF-5	0.31 <sup>b</sup>	19.3	18.5	252	128	4,5,6
MOF-177	0.21 <sup>c</sup>	28.1	27.1	250	127	7,8,9

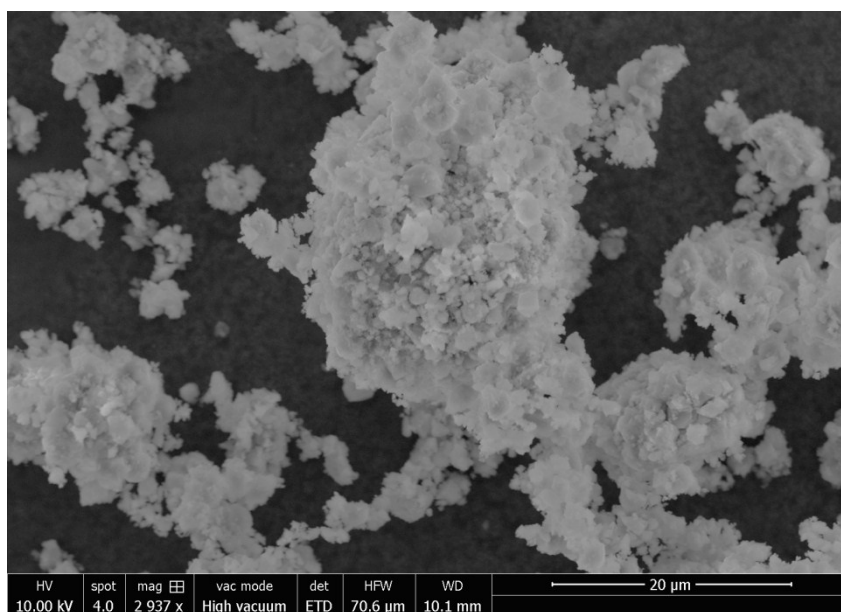
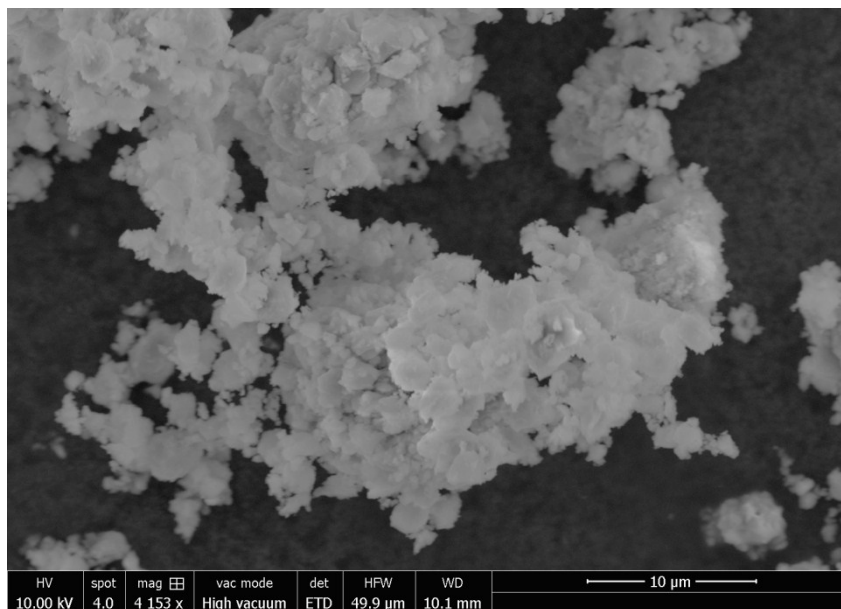
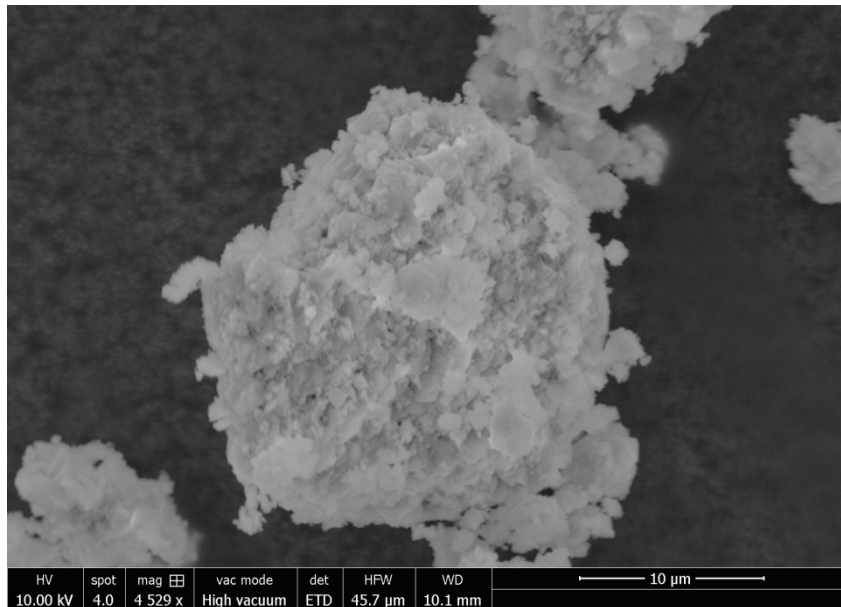
<sup>a</sup>Defined as the difference of storage capacity between 20 and 1 bar. <sup>b</sup> Packing (or pellet) density according to ref 5 and 6. <sup>c</sup> Packing density according to ref 8 and 9.

## References

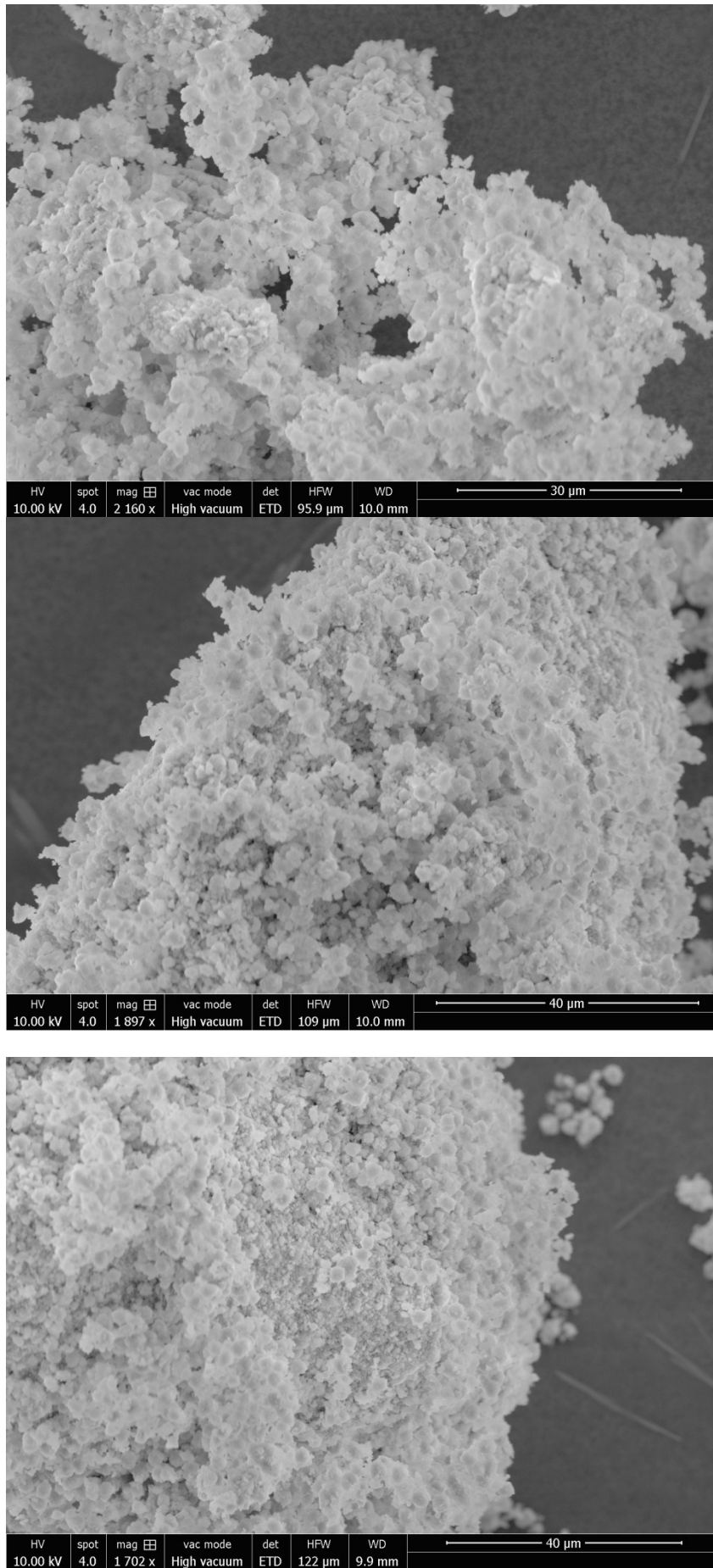
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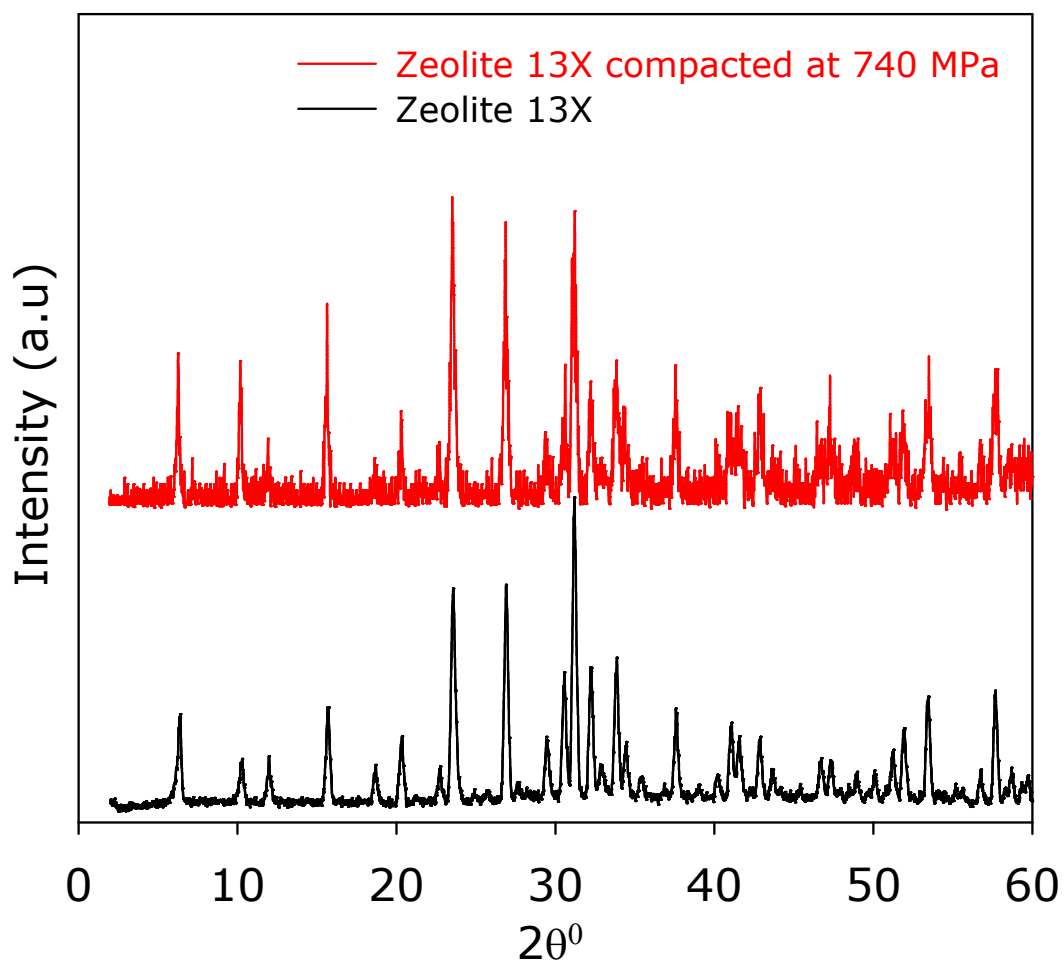
**Figure S1.** SEM images of powdered zeolite 13X



**Figure S2.** SEM images of zeolite 13X compacted at 5 tonnes (370 MPa)

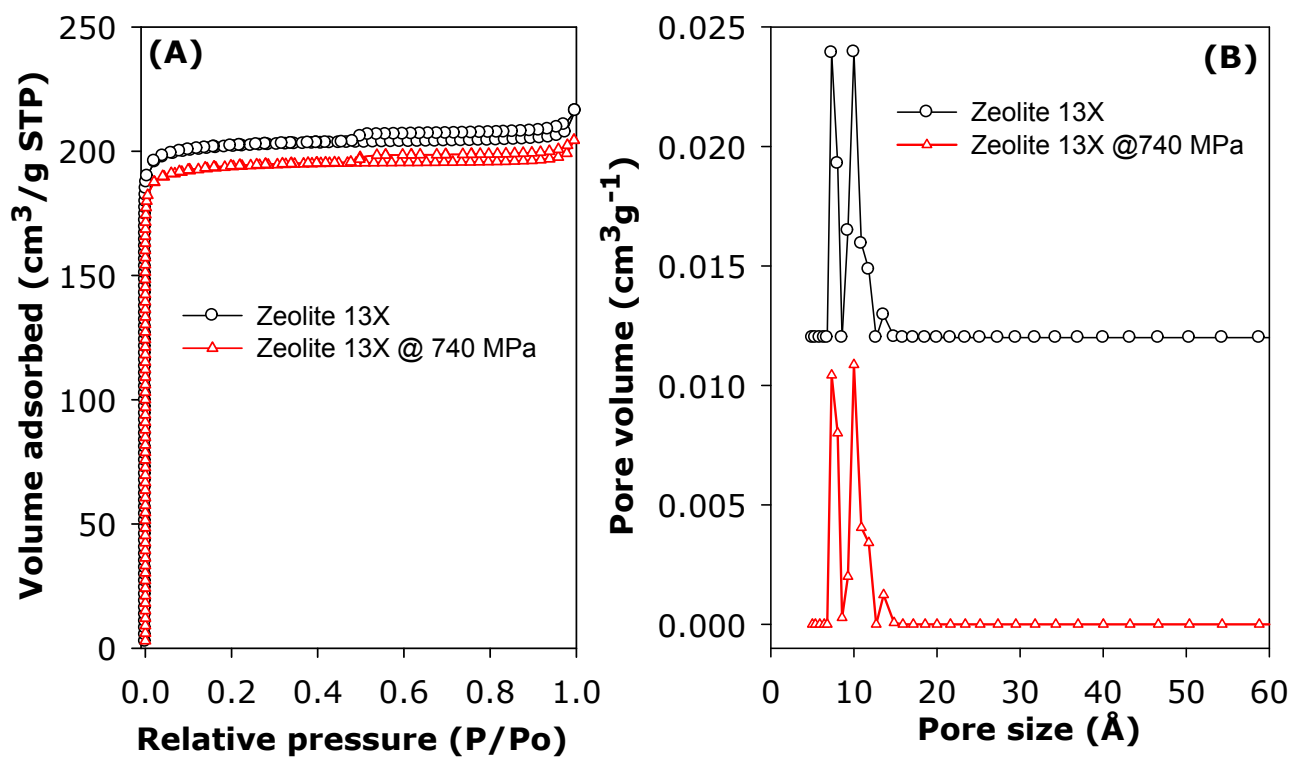


**Figure S3.** SEM images of zeolite 13X compacted at 10 tonnes (740 MPa)

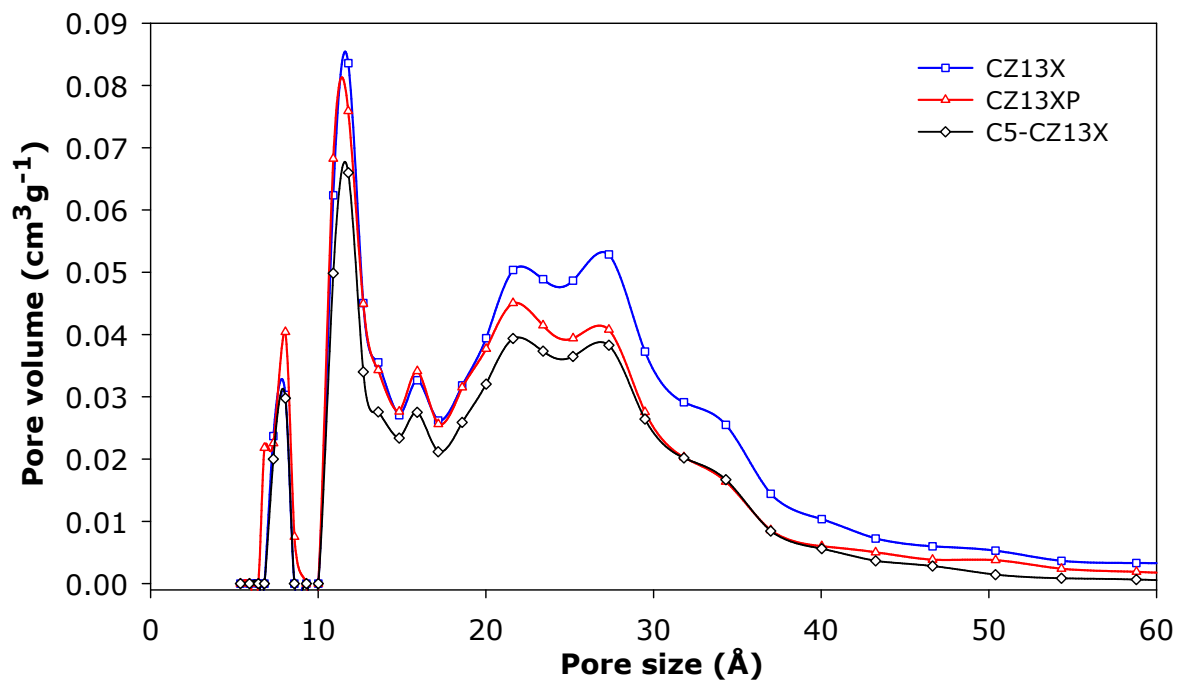


**Figure S4.** Powder XRD patterns of zeolite 13X before and after compaction and 740 MPa. The XRD patterns remain unchanged, which indicates that the structural ordering of the zeolite is preserved after compaction.

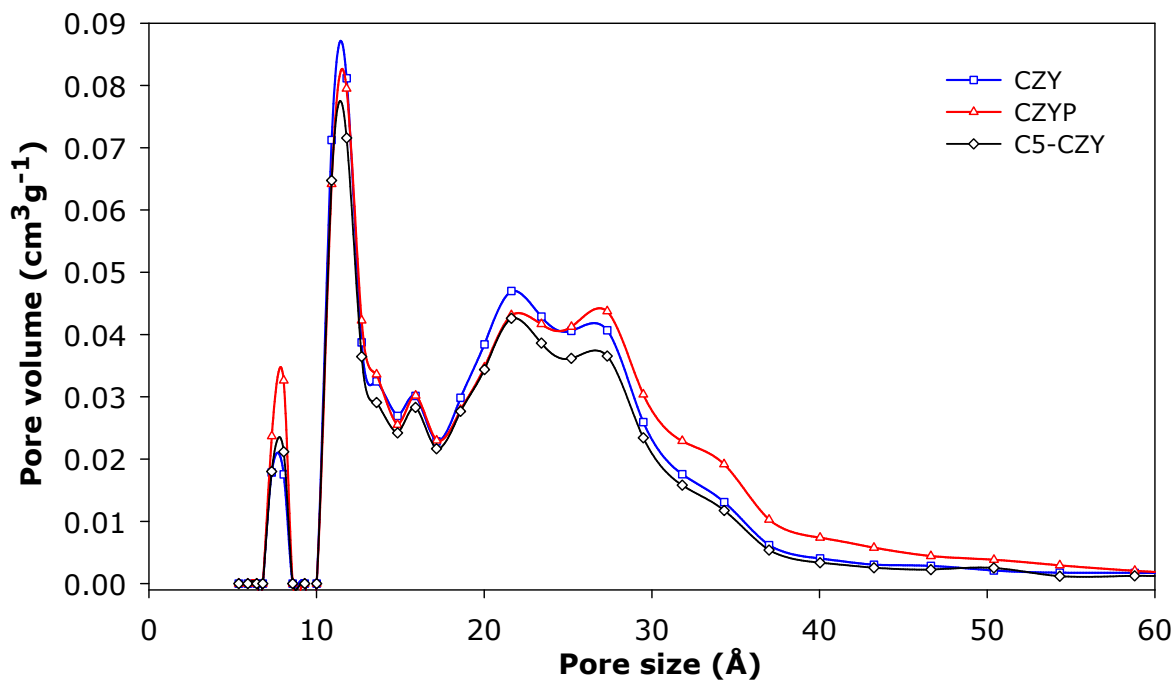




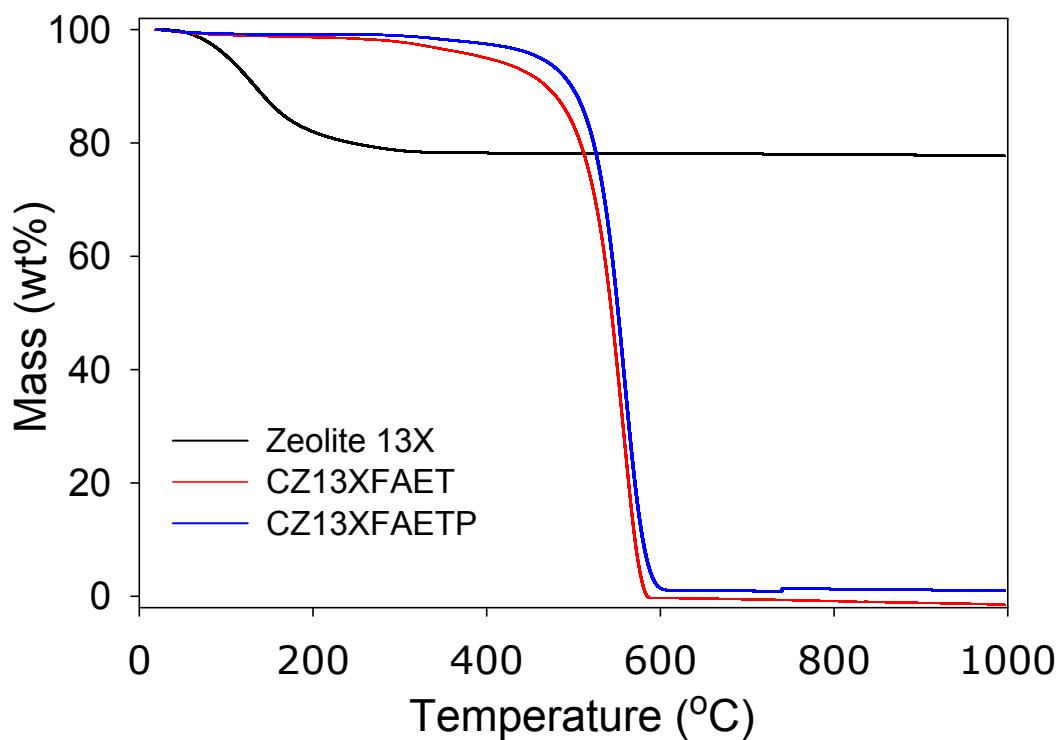
**Figure S5.** Nitrogen sorption isotherms (A) and corresponding pore size distribution (PSD) curves (B) of zeolite 13X before and after compaction at 740 MPa.



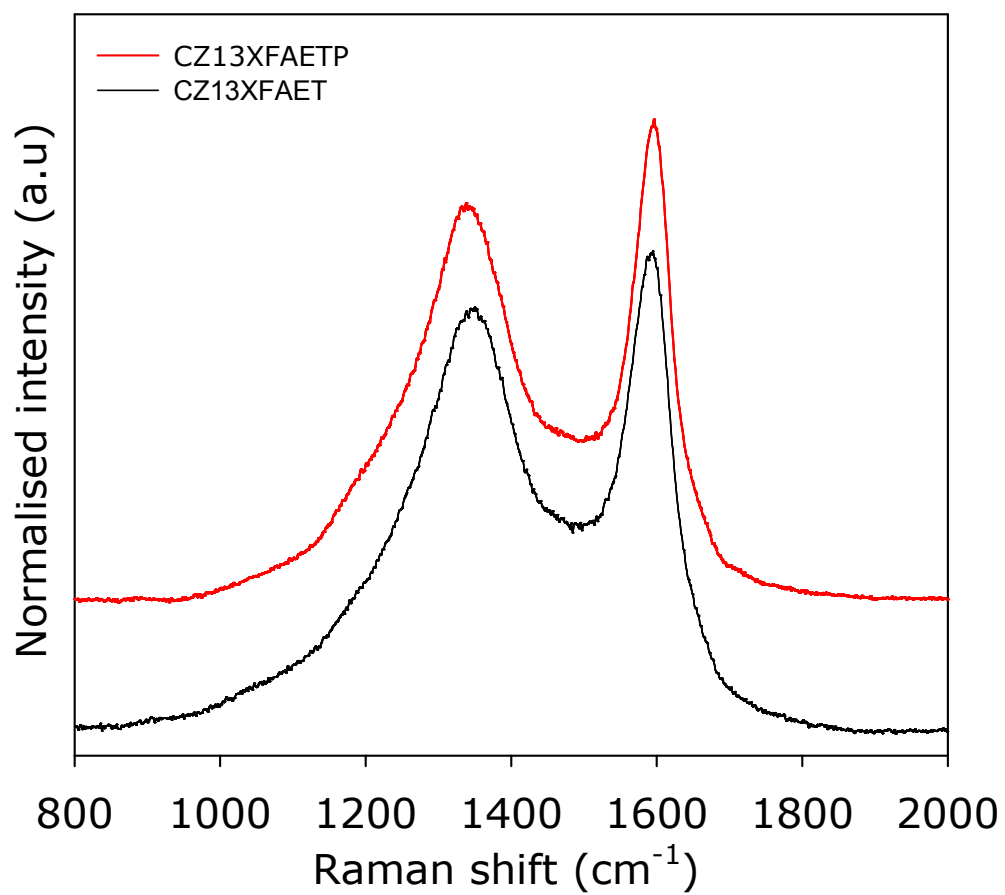
**Figure S6.** Pore size distribution curves of zeolite templated carbons prepared via a CVD route using powder (C13X) or compacted pellets (C13XP) of zeolite 13X as hard template. Sample C5-CZ13X is a directly compacted (at 370 MPa) form of CZ13X.



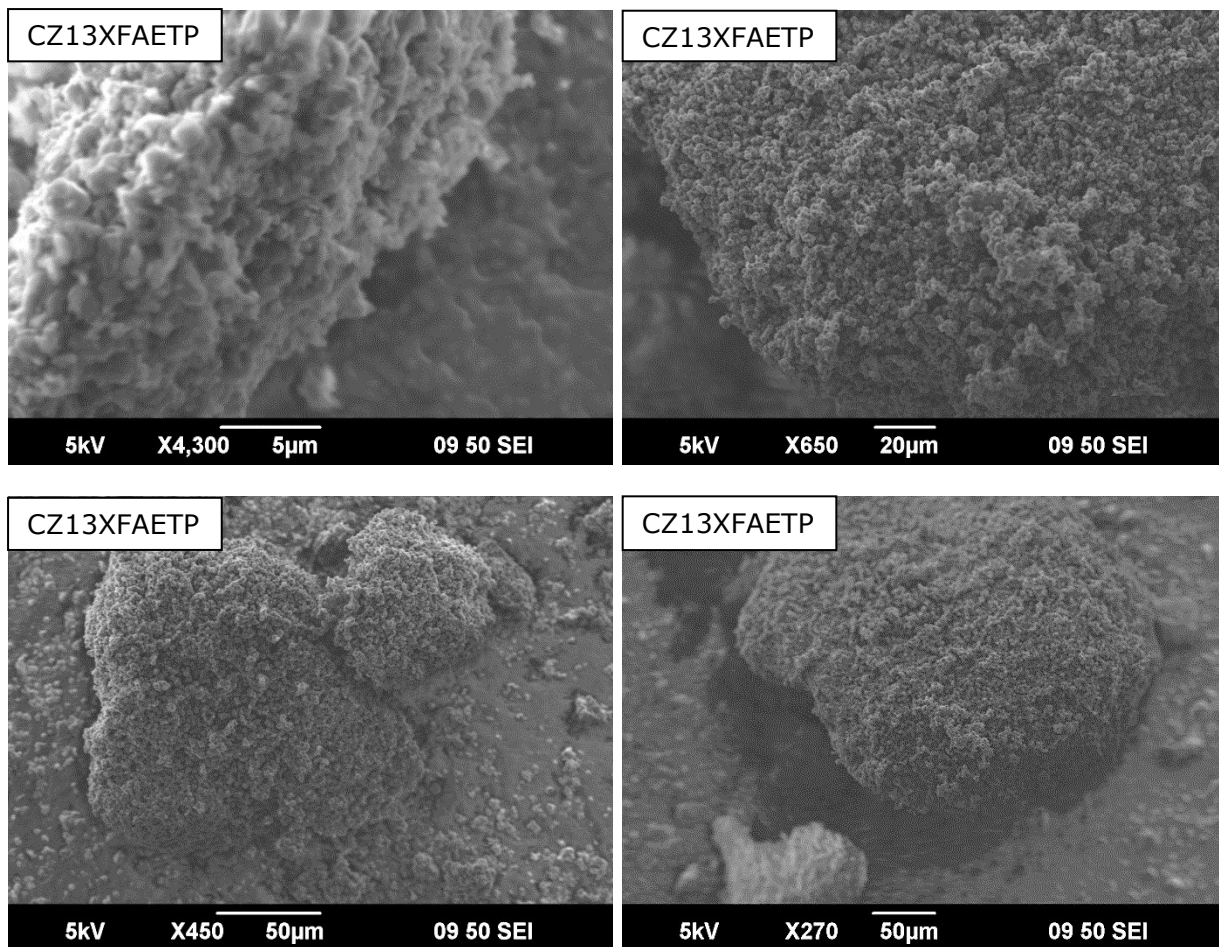
**Figure S7.** Pore size distribution curves of zeolite templated carbons prepared via a CVD route using powder (CZY) or compacted pellets (CZYP) of zeolite Y as hard template. Sample C5-CZY is a directly compacted (at 370 MPa) form of CZY.



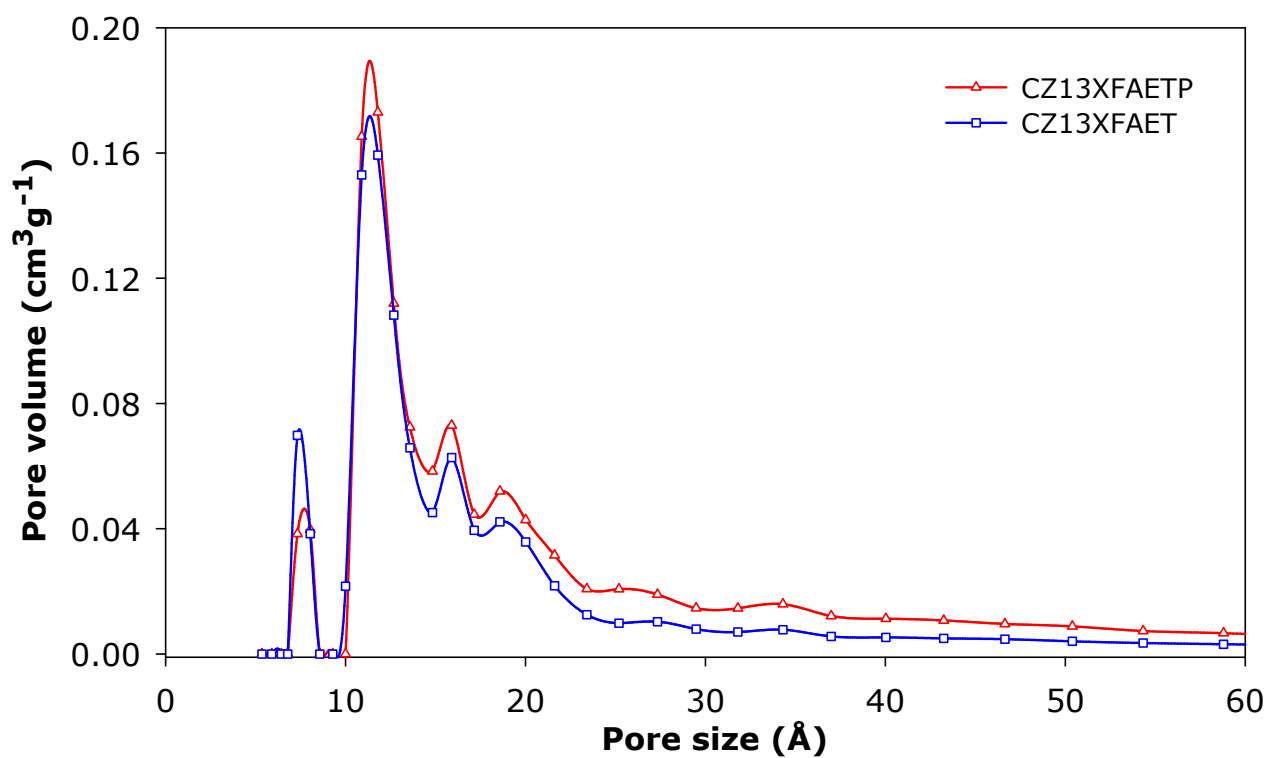
**Figure S8.** Thermogravimetric analysis (TGA) curves of zeolite 13X, and zeolite templated carbons prepared via a route that combines liquid impregnation and CVD using powder (CZ13XFAET) or compacted pellets (CZ13XFAETP) of zeolite 13X as hard template. The TGA was performed under flowing air conditions.



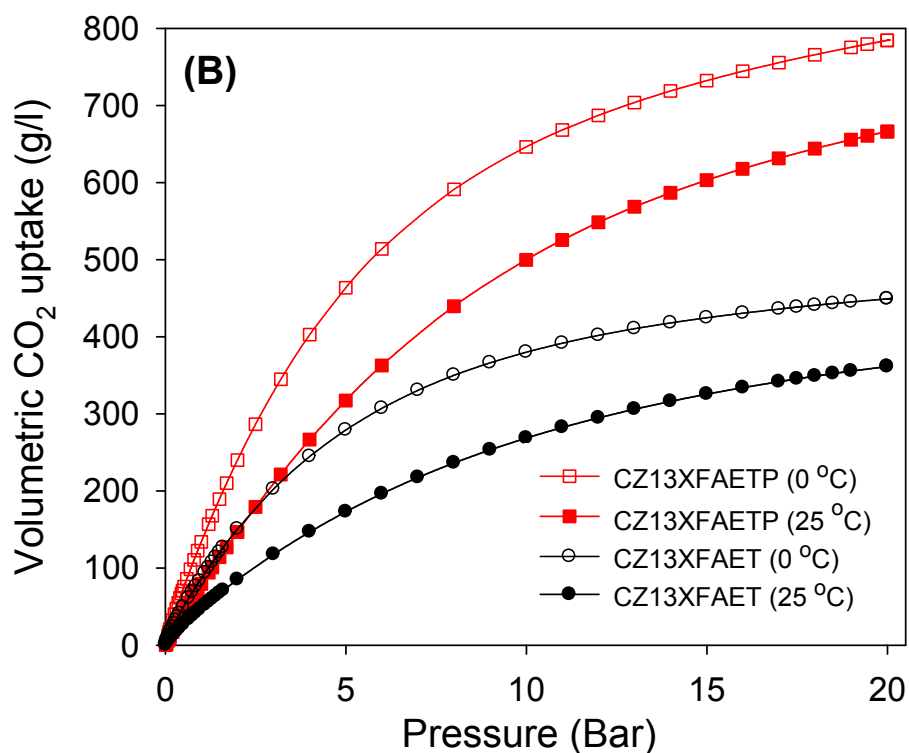
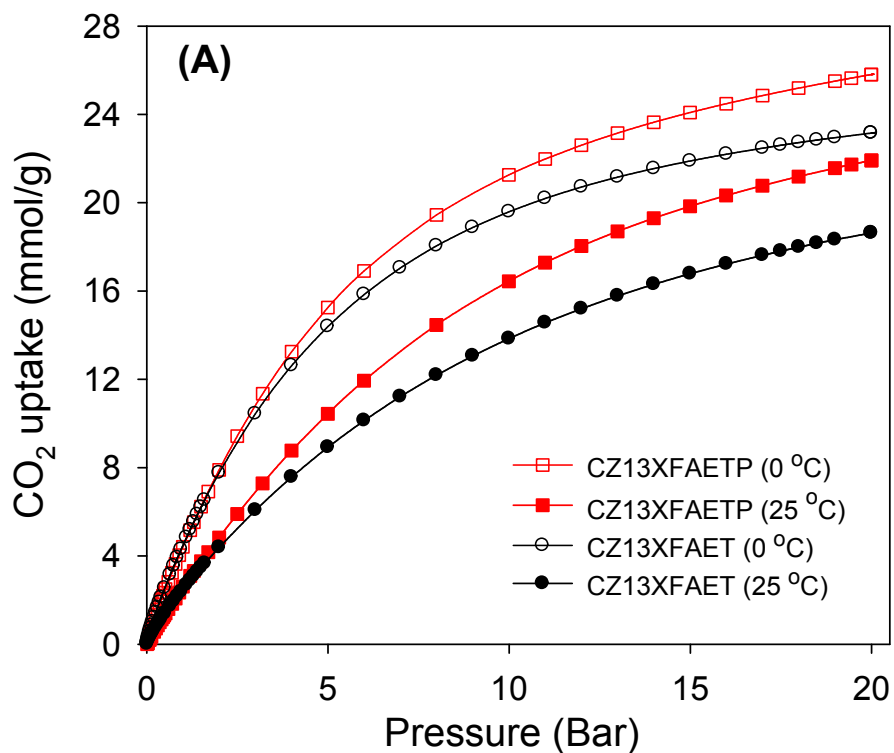
**Figure S9.** Raman spectra of zeolite templated carbons prepared via a route that combines liquid impregnation and CVD using powder (CZ13XFAET) or compacted pellets (CZ13XFAETP) of zeolite 13X as hard template.



**Figure S10.** Raman spectra of zeolite templated carbons prepared via a route that combines liquid impregnation and CVD using powder (CZ13XFAET) or compacted pellets (CZ13XFAETP) of zeolite 13X as hard template.



**Figure S11.** Pore size distribution curves of zeolite templated carbons prepared via a route that combines liquid impregnation and CVD using powder (CZ13XFAET) or compacted pellets (CZ13XFAETP) of zeolite 13X as hard template.



**Figure S12.** Gravimetric (A) and volumetric (B) CO<sub>2</sub> uptake at 0 °C or 25 °C and 0 - 20 bar for zeolite templated carbons templated by powder (CZ13XFAET) or compacted pellets (CZ13XFAETP) of zeolite 13X.