Supplementary Information

Armored Cobalt-Citrate Framework with Graphene Oxide Exhibiting Improved Thermal Stability and Selectivity for Biogas Decarburization

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Table S1 | Summary of the parameters and the enthalpies of gas adsorption on UTSA-16-GO composites at 296 and 273 K obtained from the virial equation, as shown in Figures S6~S11.

Sample	adsorbate	T/ K	A_0/ ln(mol g^{-1} Pa^{-1})	A_1/ g mol^{-1}	R^2	ΔH/ kJ mol^{-1}
UTSA-16-GO9.5	CO_2	296	-12.780±0.112	-1684.456±27.666	0.998	
		273	-11.544±0.141	-2002.903±41.121	0.998	36.1
	CH_4	296	-19.471±0.001	-420.455±4.349	0.999	
		273	-19.097±0.001	-324.566±12.874	0.994	10.9
UTSA-16-GO19	CO_2	296	-12.059±0.092	-1405.067±29.558	0.996	
		273	-10.616±0.119	-1653.615±37.188	0.998	42.1
	CH_4	296	-19.038±0.002	-296.732±7.023	0.996	
		273	-18.646±3.946E-4	-251.911±1.888	0.999	11.7
UTSA-16-GO28.5	CO_2	296	-12.021±0.075	-1943.720±56.301	0.994	
		273	-10.650±0.113	-2285.230±70.018	0.996	40.0
	CH_4	296	-19.176±6.443E-4	-407.189±8.954	0.996	
		273	-18.800±0.002	-402.117±11.438	0.996	11.4

Table S2 | Adsorption selectivities for equimolar CO_2/CH_4 mixture at 200 kPa and 296 K by IAST on different MOFs.

MOFs	T / K	S_{ads} (CO_2/CH_4)	Supplementary References
polyamine incorporated amine-MIL-101 (Cr) (**c**)	298	931	1
polyamine incorporated amine-MIL-101 (Cr) (**b**)	298	278	1
UTSA-16-GO19	296	114.4	This work
MgMOF-74	296	105	2
UTSA-16-GO9.5	296	98.7	This work
UTSA-16-GO28.5	296	89.4	This work
$[Cd_2L1(H_2O)]_2 \cdot 5H_2O$	298	66	3
UTSA-16	296	29.8	2
polyamine incorporated amine-MIL-101 (Cr) (**a**)	298	21	1
Fe_2(dobdc)	298	20.2	4
UiO-66(Zr)-CO_2H	298	19.2	5
MOF-5 (IRMOF-1)	298	15.5	6
UTSA-15a	296	14.2	2
Cu-TDPAT	296	13.8	2
bio-MOF-11	298	~12	7
$Cu(bpy\text{-}1)_2(SiF_6)$	298	~11	8
$Zn_5(BTA)_6(TDA)_2$	296	10.8	2
ZIF-78	296	10.4	2
MIL-101	296	9.6	9
UTSA-72a	293	9.6	10
UTSA-25a	296	9.4	2
UTSA-20a	296	8.3	2
$Fe_2(O_2)$(dobdc)	298	7.8	4
CuBTC	296	7.4	9
UTSA-33a	296	7.0	2
MIL-53	298	7	11
UTSA-34b	296	5.1	2
MOF-177	298	4.4	6

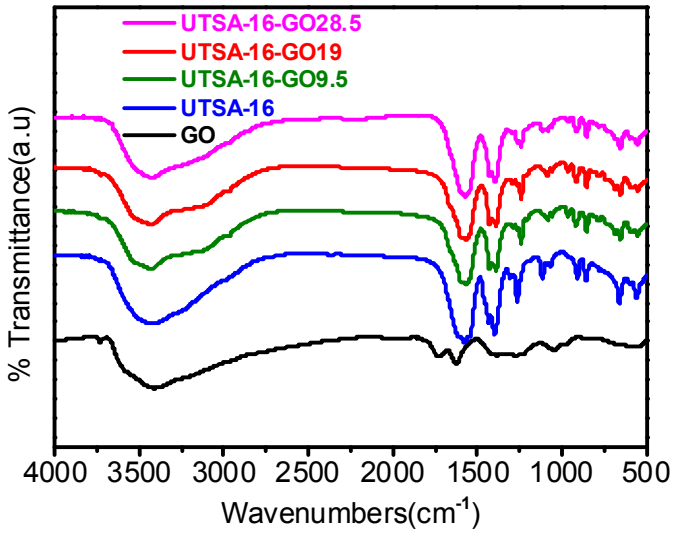


Figure S1 | FTIR spectra of as-synthesized GO, UTSA-16 and composites.

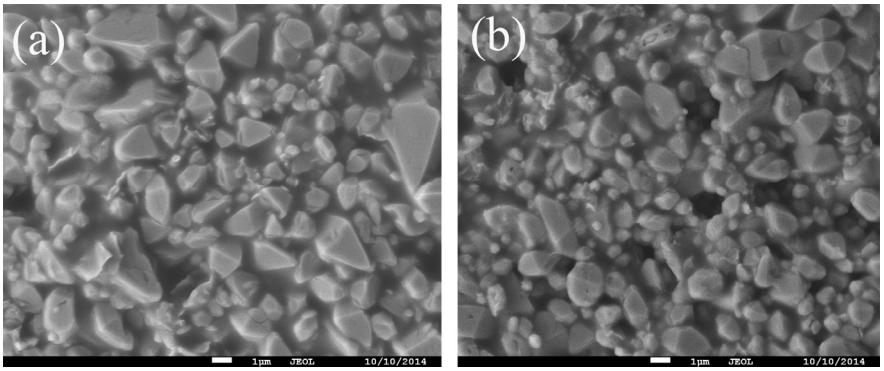


Figure S2 | SEM images for UTSA-16-GO9.5 (a) and UTSA-16-GO28.5 (b).

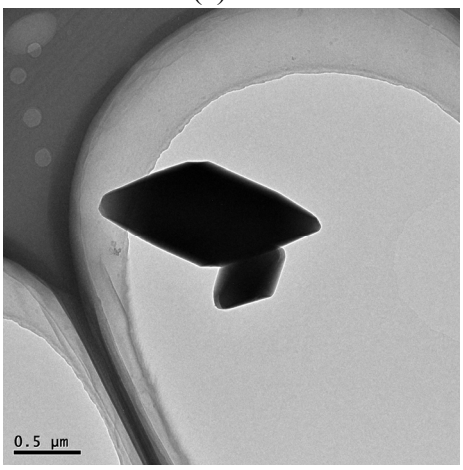


Figure S3 | TEM images for UTSA-16.

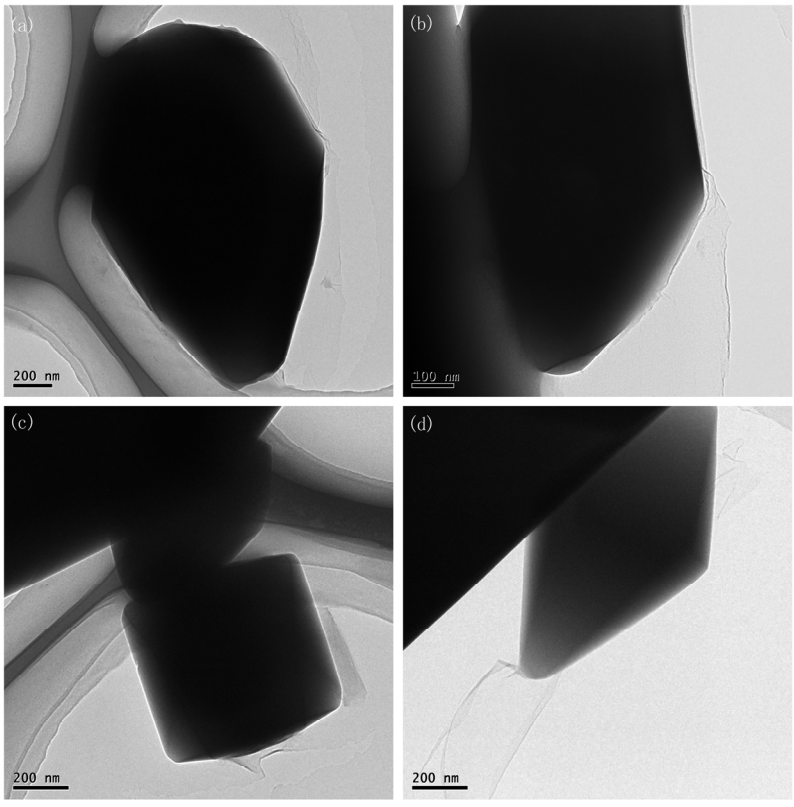


Figure S4 | TEM images for UTSA-16-GO9.5 (a, b), UTSA-16-GO28.5 (c, d).

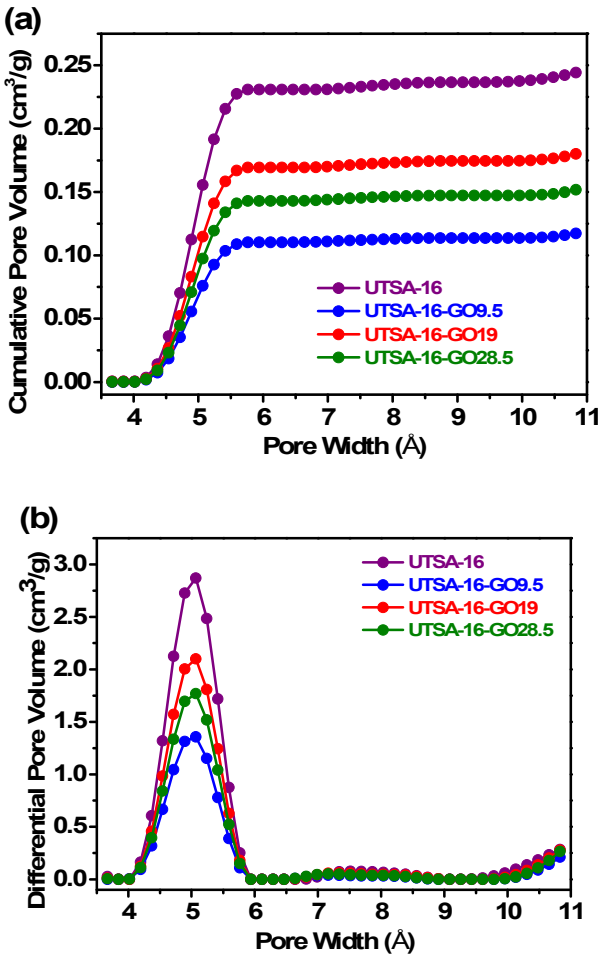


Figure S5 | (a) Cumulative pore volume and (b) pore-size distribution for UTSA-16 and its composites from CO_2 adsorption at 273 K (calculated by using a slit pore NLDFT model).

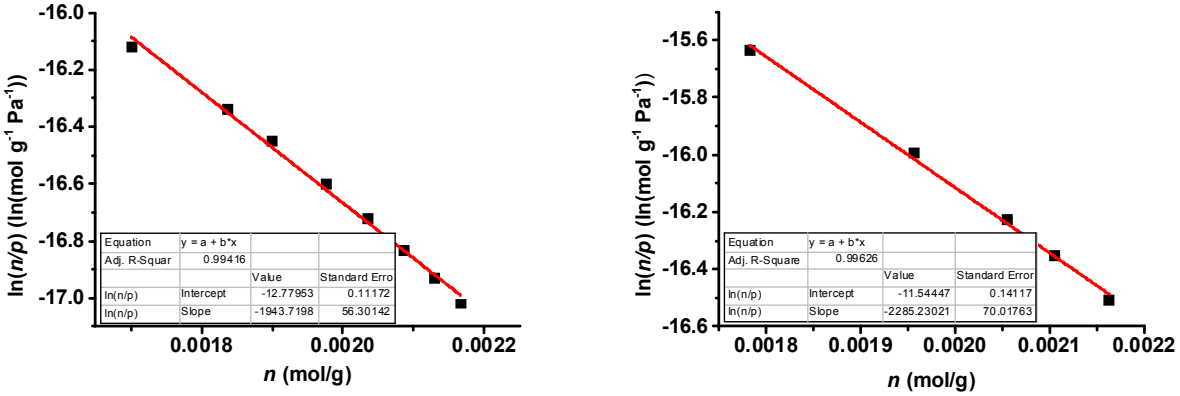


Figure S6 | The virial graphs for adsorption of CO_2 on UTSA-16-GO9.5 at 296 K (left) and 273 K (right).

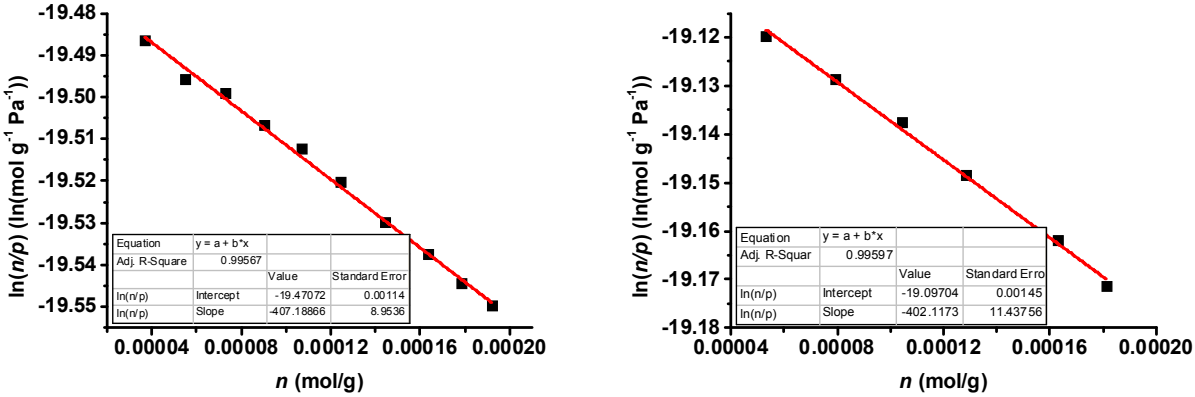
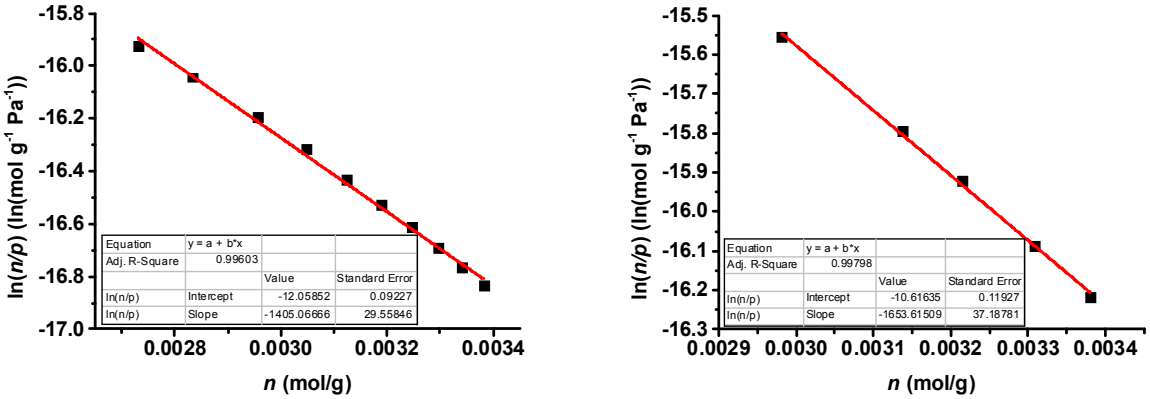


Figure S7 | The virial graphs for adsorption of CH_4 on UTSA-16-GO9.5 at 296 K (left) and 273 K (right).



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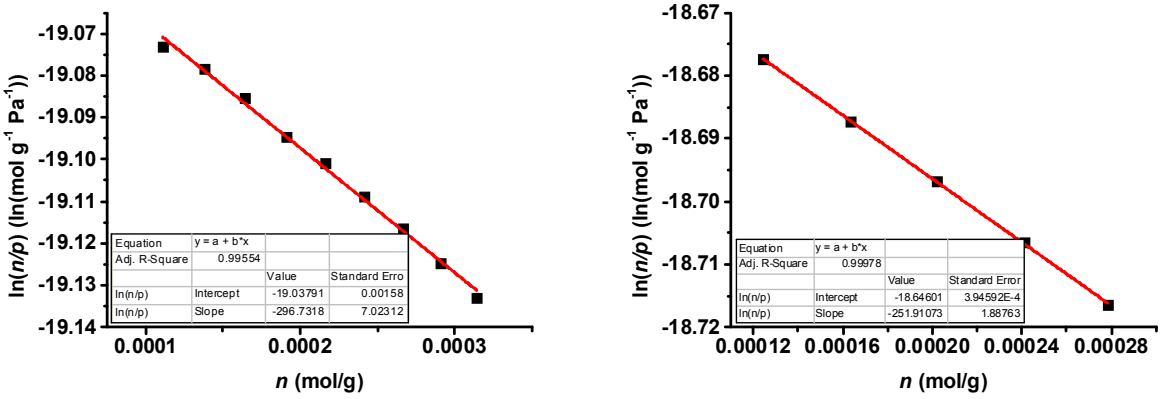


Figure S9 | The virial graphs for adsorption of CH_4 on UTSA-16-GO19 at 296 K (left) and 273 K (right).

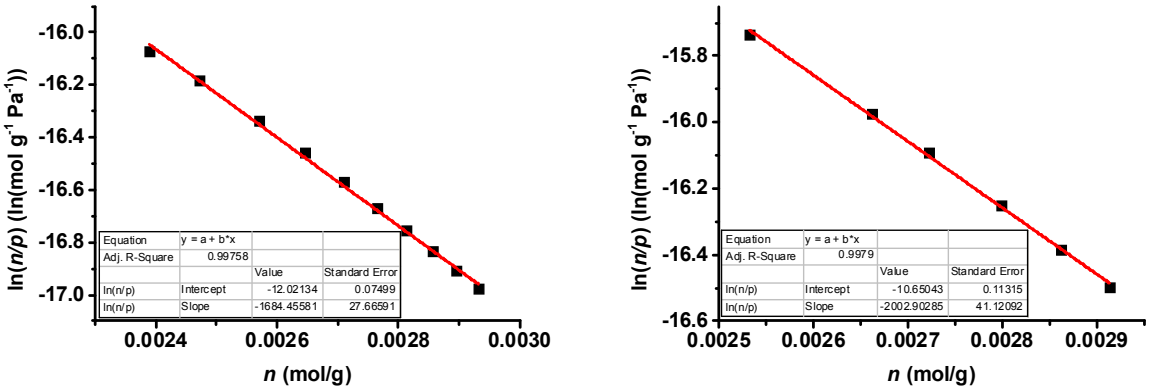
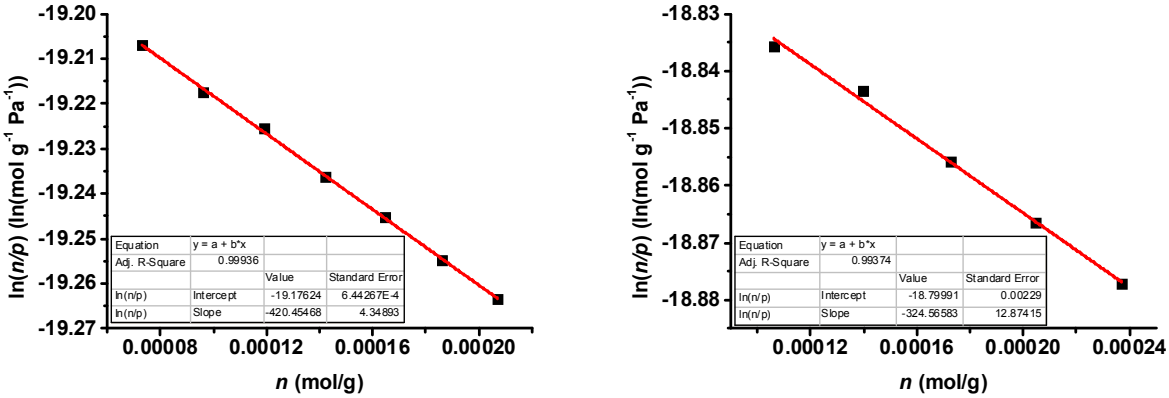


Figure S10 | The virial graphs for adsorption of CO_2 on UTSA-16-GO28.5 at 296 K (left) and 273 K (right).



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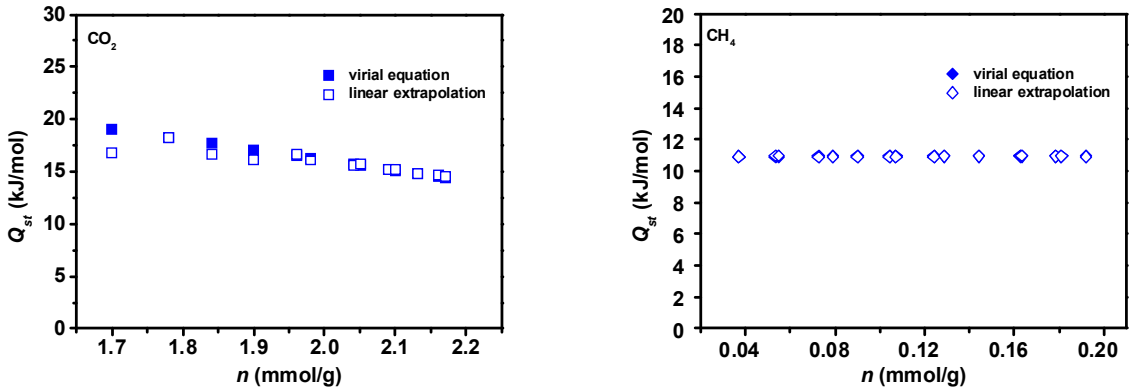

Figure S12 | Comparison of the enthalpies for gas adsorption of CO_2 (left), CH_4 (right) on UTSA-16-GO9.5 from two methods: virial equation (solid) and linear extrapolation (open).

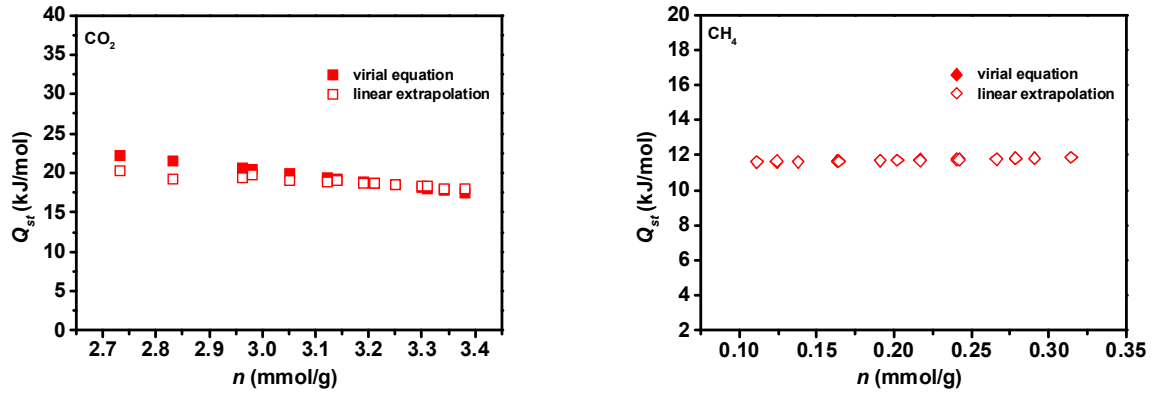

Figure S13 | Comparison of the enthalpies for gas adsorption of CO_2 (left), CH_4 (right) on UTSA-16-GO19 (magenta) from two methods: virial equation (solid) and linear extrapolation (open).

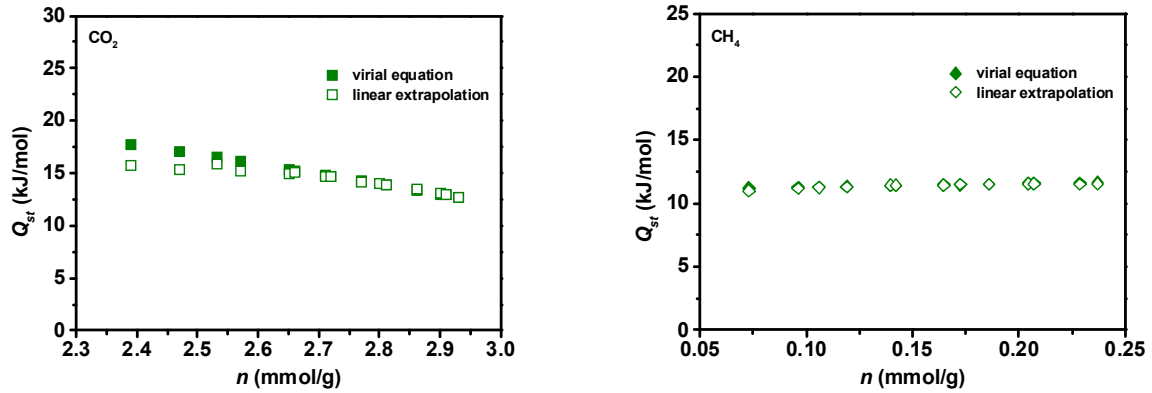

Figure S14 | Comparison of the enthalpies for gas adsorption of CO_2 (left), CH_4 (right) on UTSA-16-GO28.5 (olive) from two methods: virial equation (solid) and linear extrapolation (open).

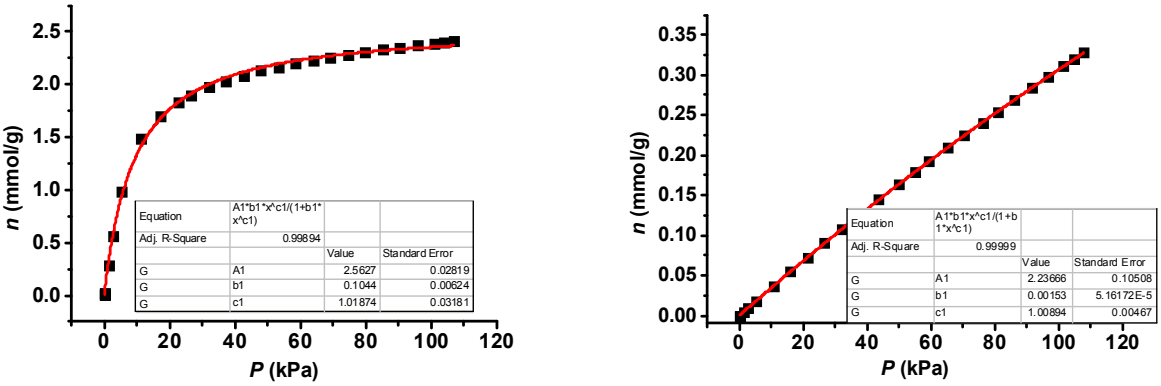

Figure S15 | The graphs of the Single-site Langmuir-Freundlich equations fit for adsorption of CO_2 (left) and CH_4 (right) on UTSA-16-GO9.5 at 296K.

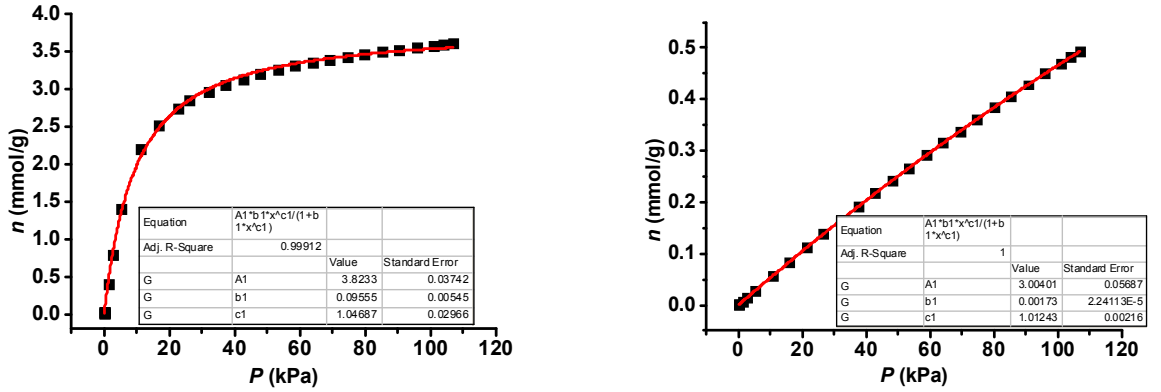

Figure S16 | The graphs of the Single-site Langmuir-Freundlich equations fit for adsorption of CO_2 (left) and CH_4 (right) on UTSA-16-GO19 at 296K.

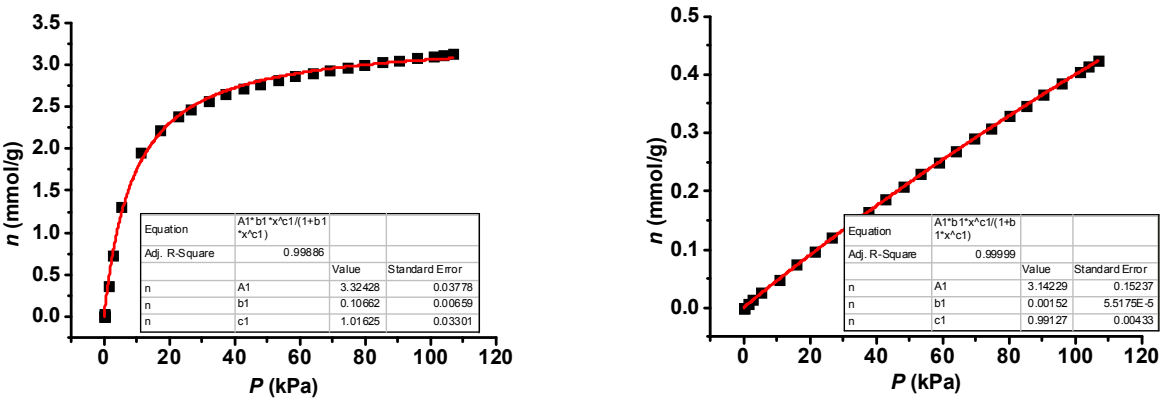

Figure S17 | The graphs of the Single-site Langmuir-Freundlich equations fit for adsorption of CO_2 (left) and CH_4 (right) on UTSA-16-GO28.5 at 296K.

Supplementary References

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