Heat Removal for Adsorbed Natural Gas Storage – Lab to Industrial Scale

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Number of tables: 5

Table S1. Physical properties of the adsorbent used in the heat removal optimization study.

Sample	AC1	AC2	AC3	AC4	AC5
ASTM mesh size	8×16	30×70	2×60	12×40	6×60
Bulk density (g/cm³)	0.47	0.39	0.49	0.54	0.50
Skeletal density (g/cm³)	2.299	2.363	2.402	2.059	2.286
Nitrogen Porosimetry (77 K)					
t-plot micropore volume (cm³/g)	0.417	0.412	0.487	0.292	0.452
t-plot micropore area (m²/g)	968.6	969.4	1186.1	722.5	1082.1
BET surface area (m²/g)	1235	1589	1426	999	1510
Total Pore Volume (cm³/g)	0.629	0.747	0.599	0.500	0.682
BJH average pore width (Å)	18.00	18.70	17.47	20.64	26.08
Results of Dubinin-Astakhov Modeling of Methane Adsorption Data at 10, 21, 38 and 56 °C.					
	AC1	AC2	AC3	AC4	AC5
Micropore volume (cm³/g)	0.380	0.449	0.416	0.2865	0.445
Micropore average pore width (Å)	9.44	10.62	9.88	10.34	9.58

In Table S1, the total pore volume from nitrogen porosimetry refers to the pores that have diameter range of 0 to 20 nm and the micropores are defined as those having a diameter range of 0 to 2 nm. For the mercury porosimetry, mesopore volume refers to the pores with a width range of 2 to 50 nm and to the macropores with a width range of 50 to 10000 nm.

Characterization. Nitrogen adsorption at 77K data is presented in Table S2. Nitrogen adsorption at 77K was used to determine the surface area and pore volume of activated carbon samples. Figure S1 shows the plot of the amount of nitrogen adsorbed versus the relative pressure. The shape of these five isotherms are of Type-I which indicates that these activated carbons are essentially microporous. After the relative pressure of 0.8, there is a slight increase in nitrogen adsorption. This is most probably due to the presence of mesopores, where the condensation of nitrogen occurs as the pressure goes up.

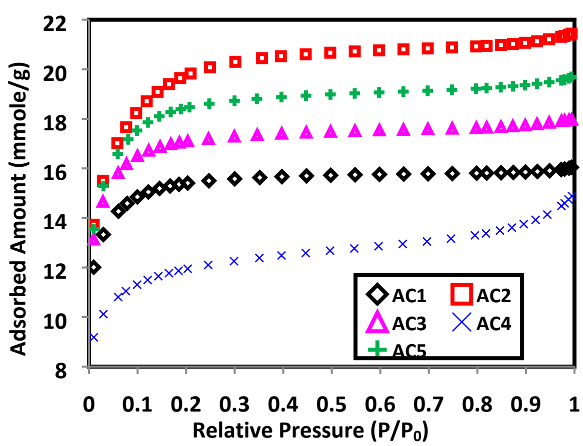

Figure S1. N_2 adsorption isotherms at 77K on granular activated carbons (data in Table S3).

Experimental Adsorption Isotherms. Figures S2.1 and S2.2 show the plots of the amount of adsorbed methane versus pressure at 21 °C for the five different activated carbons (data in Table S3). All the plots have the shape of Type-I isotherm, which also indicates that these samples are microporous materials, typical of activated carbons. In Figure 4.2 the volumetric data are based on volume to volume ratio (V/V).

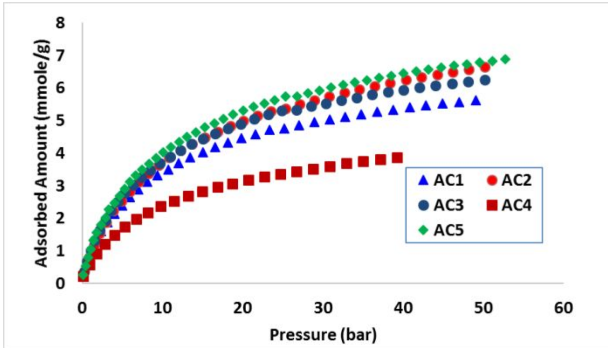

Figure S2.1. Comparison of mass to mass (mmol/g) adsorption capacity among the five granular activated carbons.

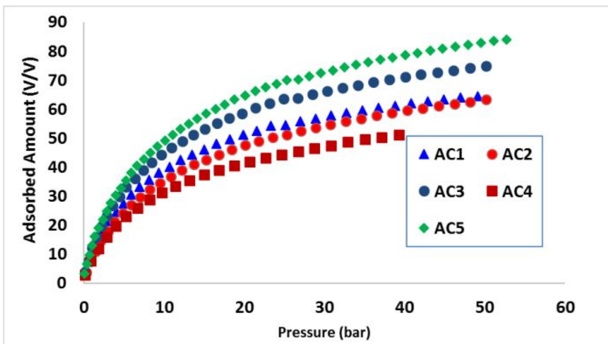

Figure S2.2. Comparison of volume to volume (v/v) adsorption capacity among the five granular activated carbons.

Based on the moles of methane adsorbed per gram of activated carbon in Figure S2.1, the capacity of methane adsorption on five activated carbons increases in the order of AC4 < AC1 < AC3 < AC2 < AC5. Although the surface areas of AC2 and AC5 are close to each other, the micropore volume of AC5 (0.452 cm^3/g) is higher than that of AC2 (0.412 cm^3/g), which is important for the adsorption on microporous adsorbents occurring by the mechanism of pore filling. This may also be due to the fact that AC5 has more slit pores that are close to the optimal pore size for methane adsorption. Earlier theoretical studies[41-43] found that for methane adsorption on slit-pore activated carbon, the optimum pore width is 11.2 to 11.4 Å in order to create the maximum density for the adsorbed phase.

Based on the volume of methane adsorbed per volume of activated carbon presented in Figure S2.2, the capacity for methane adsorption increases in the order of AC4 < AC2 < AC1 < AC3 < AC5. This change of order in comparison to the mass to mass adsorption capacity in Figure S2.1 can be attributed to the fact that the bulk density in volume to volume comparison is an important factor for volumetric adsorption capacity. The bulk density for the five activated carbons are AC1 (0.47 g/cm^3), AC2(0.39 g/cm^3), AC3(0.49 g/cm^3), AC4(0.54 g/cm^3), AC5(0.50 g/cm^3). Although AC2 has the highest BET surface area and high micropore volume, it has the lowest bulk density. Therefore, it can be concluded that the synergetic effect among BET surface area, micropore volume, micropore size distribution and bulk density play an important role in determining the volumetric adsorption capacity.

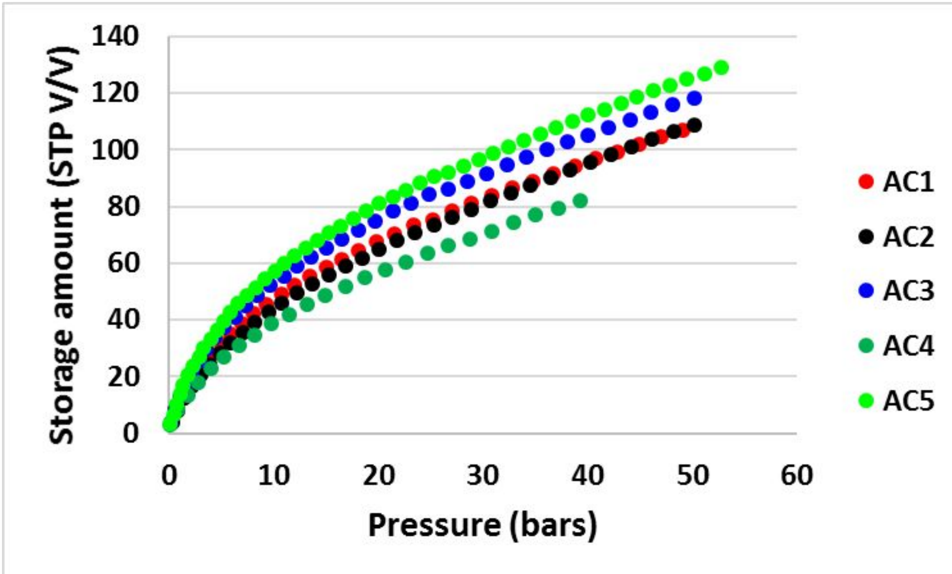

Figure S3. Comparison of volume to volume (v/v) storage capacity among the five granular activated carbons (Data in Table S3).

Table S2. Nitrogen adsorption data at 77 K for different activated carbons.

AC1		AC2		AC3		AC4		AC5	
pressure (P/P0)	adsorbed (mmole/g)	pressure (P/P0)	adsorbed (mmole/g)	pressure (P/P0)	adsorbed (mmole/g)	pressure (P/P0)	adsorbed (mmole/g)	pressure (P/P0)	adsorbed (mmole/g)
0.0095344	12.01	0.0095727	13.729	0.0094842	13.13	0.0097981	9.1837	0.0095079	13.55
0.029519	13.331	0.028714	15.503	0.028604	14.656	0.029547	10.118	0.02969	15.288
0.060606	14.268	0.058257	17.015	0.059989	15.81	0.059911	10.809	0.059125	16.585
0.078473	14.587	0.076373	17.659	0.077193	16.173	0.076021	11.048	0.08061	17.173
0.099546	14.85	0.097057	18.235	0.099733	16.496	0.09907	11.308	0.09864	17.53
0.12222	15.048	0.11882	18.708	0.12293	16.717	0.12081	11.497	0.12124	17.858
0.14525	15.191	0.14126	19.091	0.14633	16.87	0.14276	11.649	0.14463	18.101
0.16701	15.29	0.16453	19.401	0.16856	16.975	0.16461	11.774	0.16817	18.277
0.18533	15.355	0.18775	19.647	0.18623	17.04	0.18426	11.87	0.18915	18.395
0.20314	15.406	0.20923	19.829	0.20339	17.092	0.20318	11.951	0.20694	18.474
0.24699	15.5	0.24904	20.081	0.24607	17.193	0.24539	12.102	0.24761	18.61
0.3001	15.577	0.30132	20.302	0.29935	17.285	0.2986	12.257	0.29968	18.73
0.35175	15.63	0.35618	20.452	0.34655	17.348	0.35078	12.386	0.34575	18.81
0.39739	15.667	0.39519	20.529	0.3969	17.401	0.39642	12.486	0.39633	18.879
0.44804	15.698	0.44635	20.607	0.4474	17.444	0.44651	12.585	0.4469	18.936
0.49784	15.722	0.49684	20.669	0.49753	17.48	0.49647	12.678	0.49723	18.985
0.54807	15.742	0.54738	20.721	0.54783	17.51	0.54646	12.768	0.54729	19.027
0.59833	15.757	0.59745	20.766	0.59814	17.535	0.5964	12.858	0.59751	19.064
0.64826	15.77	0.6477	20.806	0.64764	17.558	0.64631	12.951	0.6475	19.1
0.69841	15.781	0.69778	20.843	0.69803	17.581	0.69606	13.05	0.69757	19.135
0.74838	15.792	0.7477	20.882	0.7481	17.604	0.74565	13.163	0.74747	19.173
0.79839	15.805	0.79748	20.924	0.79773	17.632	0.7948	13.299	0.79723	19.217
0.81923	15.812	0.819	20.946	0.81917	17.646	0.81772	13.377	0.81893	19.24
0.84884	15.823	0.84848	20.981	0.84857	17.671	0.84646	13.491	0.84821	19.277
0.8741	15.836	0.87332	21.019	0.87304	17.698	0.87104	13.609	0.87338	19.314
0.899	15.851	0.89824	21.069	0.89851	17.735	0.89554	13.752	0.89806	19.36
0.9236	15.874	0.92289	21.132	0.92281	17.782	0.91994	13.925	0.92289	19.417
0.94876	15.906	0.94763	21.213	0.94786	17.844	0.94408	14.137	0.948	19.482
0.97265	15.953	0.97255	21.311	0.97327	17.91	0.97293	14.485	0.9725	19.556
0.97969	15.973	0.97952	21.345	0.97962	17.926	0.97966	14.591	0.97937	19.584
0.98894	16.01	0.98879	21.398	0.98895	17.95	0.98765	14.748	0.9884	19.638
0.9938	16.042	0.99416	21.44	0.99432	17.965	0.99495	14.881	0.99366	19.693

Table S3. Experimental data for activated carbons methane adsorption tests at 21 °C. AC#-p is for pressure (bars), AC#-m2m is adsorbed amount (mmole/gram), AC#-v2v is adsorbed amount based on volume to volume, AC#-sv2v is stored amount based on volume to volume.

AC1-p	AC1-m2m	AC1-v2v	AC1-sv2v	AC2-p	AC2-m2m	AC2-v2v	AC2-sv2v	AC3-p	AC3-m2m	AC3-v2v	AC3-sv2v	AC4-p	AC4-m2m	AC4-v2v	AC4-sv2v	AC5-p	AC5-m2m	AC5-v2v	AC5-sv2v
0.20825	0.31741	3.6435	3.809	0.28425	0.38257	3.644	3.8811	0.16425	0.33323	3.9878	4.1184	0.12425	0.21965	2.8969	2.9884	0.09625	0.27158	3.3164	3.3914
0.62525	0.65748	7.5471	8.0444	0.82925	0.78677	7.494	8.1865	0.59625	0.6719	8.0408	8.5153	0.88925	0.57945	7.642	8.2982	0.40825	0.53451	6.5272	6.8456
1.1694	0.97751	11.221	12.152	1.4698	1.1692	11.137	12.366	1.0213	1.0129	12.122	12.935	1.8023	0.90743	11.968	13.3	0.72125	0.79799	9.7448	10.308
1.6981	1.297	14.889	16.242	2.1427	1.5465	14.73	16.524	1.562	1.3326	15.948	17.193	2.8356	1.214	16.01	18.11	1.0373	1.0597	12.94	13.75
2.363	1.5965	18.326	20.211	3.0078	1.8855	17.96	20.481	2.1988	1.6384	19.607	21.362	3.9971	1.4935	19.697	22.663	1.3697	1.3189	16.105	17.175
3.136	1.8785	21.563	24.068	3.8369	2.2297	21.238	24.46	2.8796	1.9368	23.179	25.479	5.2867	1.7428	22.985	26.917	1.7983	1.5662	19.126	20.532
4.0251	2.142	24.588	27.809	4.8782	2.5281	24.08	28.183	3.5565	2.2357	26.755	29.6	6.6885	1.97	25.982	30.969	2.3389	1.7941	21.908	23.739
4.9663	2.3957	27.5	31.481	5.8955	2.8327	26.981	31.949	4.4336	2.5038	29.964	33.516	8.1784	2.1783	28.729	34.843	2.8636	2.0253	24.732	26.976
5.8875	2.6535	30.459	35.186	7.0209	3.114	29.661	35.59	5.2707	2.7767	33.229	37.459	9.7645	2.3658	31.202	38.523	3.3162	2.2673	27.687	30.287
6.9569	2.8889	33.162	38.758	8.2145	3.3826	32.219	39.17	6.3361	3.0124	36.05	41.144	11.431	2.5375	33.466	42.062	3.969	2.4779	30.259	33.374
8.0983	3.1111	35.711	42.239	9.4801	3.6335	34.609	42.649	7.3814	3.251	38.906	44.852	13.173	2.6905	35.484	45.42	4.6259	2.6891	32.839	36.473
9.3199	3.3201	38.112	45.641	10.814	3.8675	36.839	46.032	8.4868	3.4782	41.625	48.475	14.971	2.8306	37.331	48.661	5.2787	2.8996	35.409	39.561
10.706	3.5007	40.184	48.854	12.208	4.0917	38.974	49.378	9.6843	3.6929	44.194	52.027	16.829	2.9551	38.973	51.751	5.8915	3.1159	38.051	42.69
12.023	3.6948	42.413	52.173	13.71	4.2922	40.883	52.599	10.942	3.8959	46.624	55.494	18.732	3.0707	40.498	54.768	6.6364	3.3147	40.478	45.711
13.469	3.8664	44.382	55.344	15.272	4.4756	42.63	55.716	12.26	4.0887	48.931	58.893	20.678	3.1774	41.905	57.712	7.4575	3.4989	42.727	48.616
15.019	4.0203	46.149	58.406	16.834	4.6597	44.383	58.848	13.605	4.275	51.16	62.242	22.657	3.2739	43.178	60.558	8.3186	3.6783	44.918	51.497
16.533	4.178	47.959	61.488	18.444	4.8327	46.032	61.926	15.051	4.4471	53.22	65.511	24.672	3.3597	44.309	63.302	9.1837	3.8569	47.099	54.374
18.155	4.3204	49.593	64.492	20.098	4.9953	47.58	64.95	16.565	4.606	55.122	68.685	26.71	3.4422	45.397	66.032	10.093	4.0272	49.178	57.186
19.837	4.4507	51.089	67.417	21.796	5.149	49.045	67.939	18.127	4.7563	56.921	71.805	28.761	3.5188	46.408	68.707	11.05	4.1913	51.182	59.965
21.564	4.573	52.493	70.296	23.53	5.2904	50.392	70.851	19.749	4.896	58.592	74.854	30.835	3.5887	47.33	71.323	12.035	4.3495	53.114	62.697
23.338	4.7243	54.229	73.558	25.296	5.376	51.207	73.27	21.415	5.0598	60.552	78.238	32.926	3.6921	48.693	74.406	13.065	4.5006	54.96	65.381
25.152	4.7519	54.546	75.444	27.095	5.5068	52.452	76.159	23.138	5.1844	62.043	81.209	35.033	3.7557	49.532	76.988	14.142	4.6466	56.743	68.045
27.003	4.8541	55.719	78.227	28.917	5.6311	53.636	79.017	24.884	5.304	63.475	84.151	37.156	3.8141	50.303	79.528	15.247	4.7855	58.438	70.647
28.893	4.9486	56.805	80.967	30.771	5.7471	54.741	81.836	26.674	5.3333	63.825	86.058	39.266	3.8735	51.086	82.082	16.437	4.9121	59.985	73.175
30.811	5.0375	57.825	83.677	32.642	5.8582	55.8	84.635	28.504	5.437	65.066	88.901					17.566	5.0463	61.624	75.748
32.762	5.1199	58.771	86.353	34.532	5.9636	56.803	87.407	30.371	5.5336	66.223	91.7					18.78	5.1923	63.406	78.538
34.748	5.1953	59.636	88.989	36.443	6.0632	57.752	90.154	32.261	5.6252	67.319	94.47					20.082	5.3028	64.756	80.974
36.751	5.2668	60.457	91.608	38.373	6.157	58.645	92.876	34.18	5.7109	68.345	97.206					21.351	5.4179	66.161	83.443
38.762	5.3358	61.249	94.217	40.312	6.2476	59.508	95.586	36.122	5.7915	69.309	99.912					22.661	5.5262	67.484	85.868
40.8	5.399	61.974	96.794	42.266	6.3332	60.324	98.274	38.073	5.8697	70.245	102.61					23.991	5.6309	68.763	88.27
42.859	5.4576	62.647	99.35	44.225	6.4165	61.118	100.96	40.043	5.9433	71.126	105.28					25.34	5.732	69.997	90.651
44.905	5.5171	63.33	101.92	46.203	6.494	61.855	103.61	42.034	6.0124	71.952	107.92					26.734	5.7474	70.185	92.027
46.98	5.5707	63.945	104.45	48.182	6.57	62.58	106.26	44.048	6.0762	72.716	110.53					28.136	5.8392	71.306	94.35
49.083	5.6183	64.492	106.96	50.172	6.642	63.265	108.9	46.075	6.1369	73.442	113.13					29.562	5.9267	72.375	96.646
								48.11	6.195	74.138	115.72					31.012	6.0098	73.39	98.915
								50.14	6.2527	74.828	118.31					32.482	6.0892	74.359	101.16
																33.963	6.1661	75.297	103.39
																35.465	6.2391	76.19	105.6
																36.979	6.3097	77.051	107.8
																38.509	6.377	77.873	109.98
																40.043	6.4431	78.68	112.15
																41.601	6.5047	79.432	114.29
																43.163	6.565	80.169	116.43
																44.733	6.6234	80.882	118.56
																46.315	6.6791	81.563	120.67
																47.913	6.7317	82.205	122.77
																49.507	6.7843	82.847	124.87
																51.113	6.8342	83.457	126.95
																52.727	6.8823	84.044	129.03

Table S4. Experimental and model data for Figure 3 (AC5-methane system).

10 °C		21 °C		38 °C		56 °C	
Presure (bar)	Adsorbed amount (mmole/g)	Presure (bar)	Adsorbed amount (mmole/g)	Presure (bar)	Adsorbed amount (mmole/g)	Presure (bar)	Adsorbed amount (mmole/g)
0.06025	0.30914	0.09625	0.27157	0.16425	0.19688	0.30825	0.20968
0.26425	0.58733	0.40825	0.53448	0.48025	0.46026	0.73725	0.45941
0.56825	0.85126	0.72125	0.79794	0.90125	0.70965	1.2816	0.6913
0.88925	1.1115	1.0373	1.0596	1.3577	0.9535	1.9224	0.91072
1.2095	1.3717	1.3697	1.3188	1.9064	1.1839	2.4711	1.142
1.5379	1.6319	1.7983	1.5661	2.359	1.427	3.156	1.3552
1.9585	1.8778	2.3389	1.7939	2.9998	1.6437	3.8889	1.5614
2.387	2.1211	2.8636	2.0251	3.6366	1.8611	4.726	1.7512
2.8316	2.3646	3.3162	2.2671	4.3135	2.0711	5.5551	1.9433
3.2882	2.6035	3.969	2.4776	5.0384	2.2753	6.4162	2.1296
3.8489	2.8263	4.6259	2.6888	5.8675	2.4626	7.3293	2.3095
4.4216	3.0496	5.2787	2.8992	6.6725	2.6534	8.2866	2.483
5.0584	3.2591	5.8915	3.1155	7.5296	2.839	9.2718	2.6522
5.6632	3.4748	6.6364	3.3142	8.4227	3.0163	10.301	2.8124
6.3561	3.6792	7.4575	3.4983	9.3599	3.1885	11.383	2.9688
7.089	3.8758	8.3186	3.6776	10.333	3.355	12.448	3.1248
7.9021	4.0598	9.1837	3.8561	11.342	3.5146	13.641	3.2634
8.7191	4.2417	10.093	4.0263	12.4	3.6683	14.831	3.4024
9.5842	4.4158	11.05	4.1904	13.497	3.8144	16.097	3.5289
10.493	4.5854	12.035	4.3485	14.575	3.9633	17.346	3.6588
11.439	4.7464	13.065	4.4995	15.792	4.0936	18.636	3.7813
12.424	4.8995	14.142	4.6454	16.986	4.2247	19.949	3.9001
13.449	5.0485	15.247	4.7841	18.175	4.3573	21.311	4.0143
14.515	5.1909	16.437	4.9107	19.489	4.4711	22.689	4.1233
15.72	5.3094	17.566	5.0448	20.783	4.5861	24.095	4.2293
16.805	5.4394	18.78	5.1906	22.116	4.7011	25.529	4.3041
17.983	5.5651	20.082	5.301	23.474	4.8094	26.986	4.4042
19.249	5.6762	21.351	5.416	24.852	4.9143	28.46	4.5017
20.482	5.7919	22.661	5.5242	26.266	5.0134	29.966	4.5942
21.764	5.8993	23.991	5.6287	27.695	5.1096	31.488	4.6841
23.057	6.0042	25.34	5.7297	29.157	5.2006	33.03	4.7707
24.387	6.1027	26.734	5.7449	30.631	5.2894	34.596	4.8535
25.741	6.1967	28.136	5.8365	32.121	5.3753	36.17	4.9348
27.115	6.2868	29.562	5.9239	33.647	5.4553	37.776	5.011
28.516	6.3717	31.012	6.0068	35.181	5.5336	39.394	5.0852
29.942	6.4519	32.482	6.086	36.735	5.6084	41.012	5.1591
31.368	6.5314	33.963	6.1626	38.297	5.6815	42.655	5.2291
32.83	6.6043	35.465	6.2355	39.875	5.7517	44.301	5.2982
34.3	6.6752	36.979	6.3058	41.477	5.7395	45.963	5.3646
35.794	6.7412	38.509	6.373	43.079	5.8051	47.641	5.4283
37.304	6.8039	40.043	6.4389	44.697	5.8678	49.319	5.4917
38.814	6.8658	41.601	6.5002	46.311	5.9307	51.009	5.5531
40.352	6.8439	43.163	6.5603	47.941	5.9906	52.703	5.6135
41.886	6.9001	44.733	6.6185	49.575	6.0495		
43.428	6.9542	46.315	6.674	51.218	6.1067		
44.994	7.0034	47.913	6.7263	52.876	6.1609		
46.552	7.0532	49.507	6.7786	54.542	6.2135		
48.13	7.0988	51.113	6.8284				
49.708	7.1435	52.727	6.8761				
51.282	7.1881						

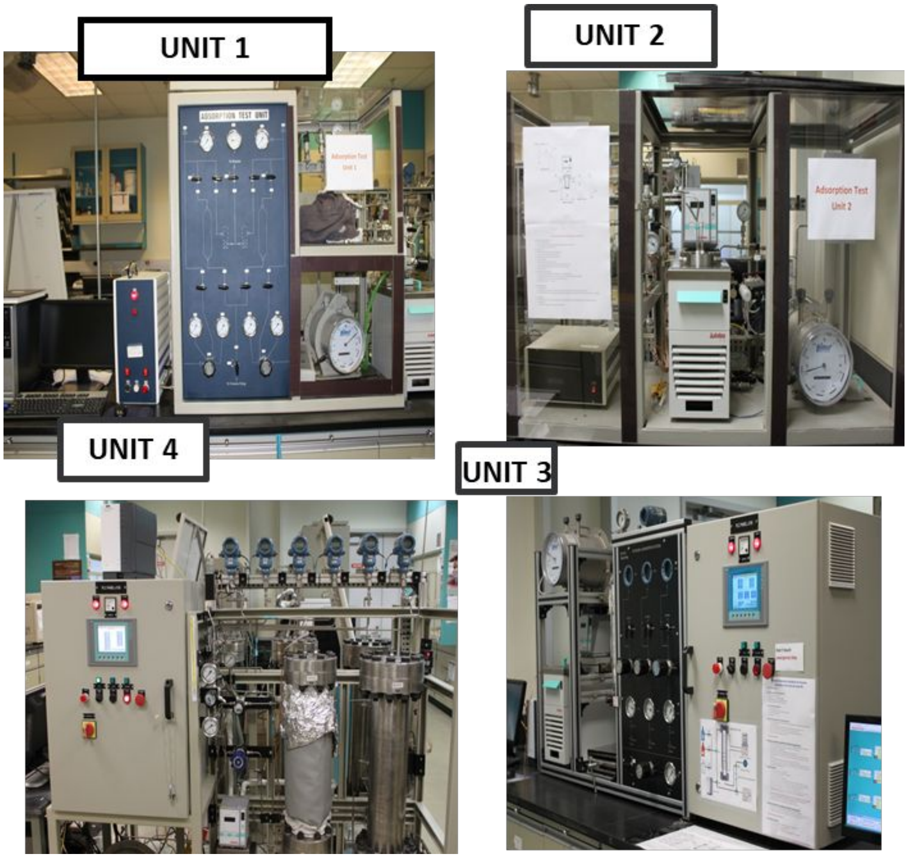

Figure s4. Pictorial view of gas adsorption lab setup.

Table S5. Activated carbon adsorbents tested for natural gas vehicles tanks reported in literature.

Project	Investigation Method	Pressure (bar)	Tank Uptake (volume to volume)
Atlanta Gas Light Adsorbent Research Group	Chrysler B Van, Dodge Dakota truck	35-40	150 in laboratory conditions
EU FPS LEVINGS program	FIAT Marea on board, field testing	35-40	123
Oak Ridge National Laboratory	Lab Investigation	35	150
Honda Motors	Tank Development Adsorbent Lab Test	35	155
University of Petroleum, China	Car Xiali 7131U on board field testing	50	100-110
University of Petroleum, China	Car Xiali 7131U on board field testing	125	170-180
Brazilian Gas Technology Center	Lab investigation on full size prototype	35-40	130-150

ALL-CRAFT (University of Missouri	Lab investigation, adsorbent optimization, field testing	35	Intragranular capability: 202; Monolith capability: 161
ALL-CRAFT (University of Missouri	Lab investigation, adsorbent optimization, field testing	250	Intragranular capability: 337; Monolith capability: to be determined