Supporting Information for:

Idealized Carbon-Based Materials Exhibiting Record Deliverable Capacities for Vehicular Methane Storage

Sean P. Collins[a], Eric Perim[c], Tom D. Daff[a], Munir S. Skaf[b,*], Douglas S. Galvão[c,*] and Tom K. Woo[a,*]

[a]Centre for Catalysis Research and Innovation, Department of Chemistry and Biomolecular Science, University of Ottawa, 10 Marie Curie Private, Ottawa K1N 6N5, Canada

[b]Institute of Chemistry, University of Campinas, Cx. P. 6154, Campinas, SP 13084-862, Brazil

[c]Applied Physics Department, University of Campinas, Campinas, SP 13083-970, Brazil

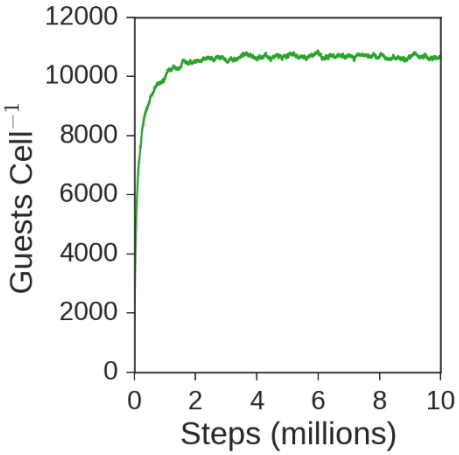

Figure S1. Number of CH_4 guest molecules in grand canonical Monte Carlo cell of an isolated graphene nanoscroll 20 Å interlayer spacing and 3000 Å in length at 298 K and 65 bar of CH_4 as a function of the steps. This is the most highly adsorbing structure based on number of guest molecules, and by the end of the equilibration period (10 million steps), equilibrium is reached.

Further GCMC Simulation Details. A Lennard-Jones cut-off distances of 12.5 Å was used for all simulations, and supercells were constructed for each structure that satisfies the minimum image criterion.

Isosteric heats of adsorbtion were calculated using Equation $+RT$ S1

$$q_{st} = -\frac{\langle EN\rangle - \langle E\rangle\langle N\rangle}{\langle N^2\rangle - \langle N\rangle^2} + RT \quad \text{S1}$$

In this equation q_{st} is the isosteric heat of adsorbtion, E is total energy of the guest molecules in the simulations, N is the number of guest of guest molecules, R is the gas constant, and T is the temperature. The angle brackets denote the average in the grand ensemble.

Table S1. Heats of adsorption of CH_4 in studied Schwarzites that had non-zero adsorption, at the adsorption condition of 298 K and 65 bar. Errors are windowed standard deviation from GCMC.

Schwarzite	Heat of Adsorption (kJ/mol)
C168	21.4 +/- 0.3
G8bal	24.9 +/- 0.4
D766	21.8 +/- 0.4
D8bal	24.7 +/- 0.4
P688	38.0 +/- 0.4
P8bal	29.3 +/- 0.6
IWPg	23.2 +/- 0.7
P7par	25.1 +/- 0.7

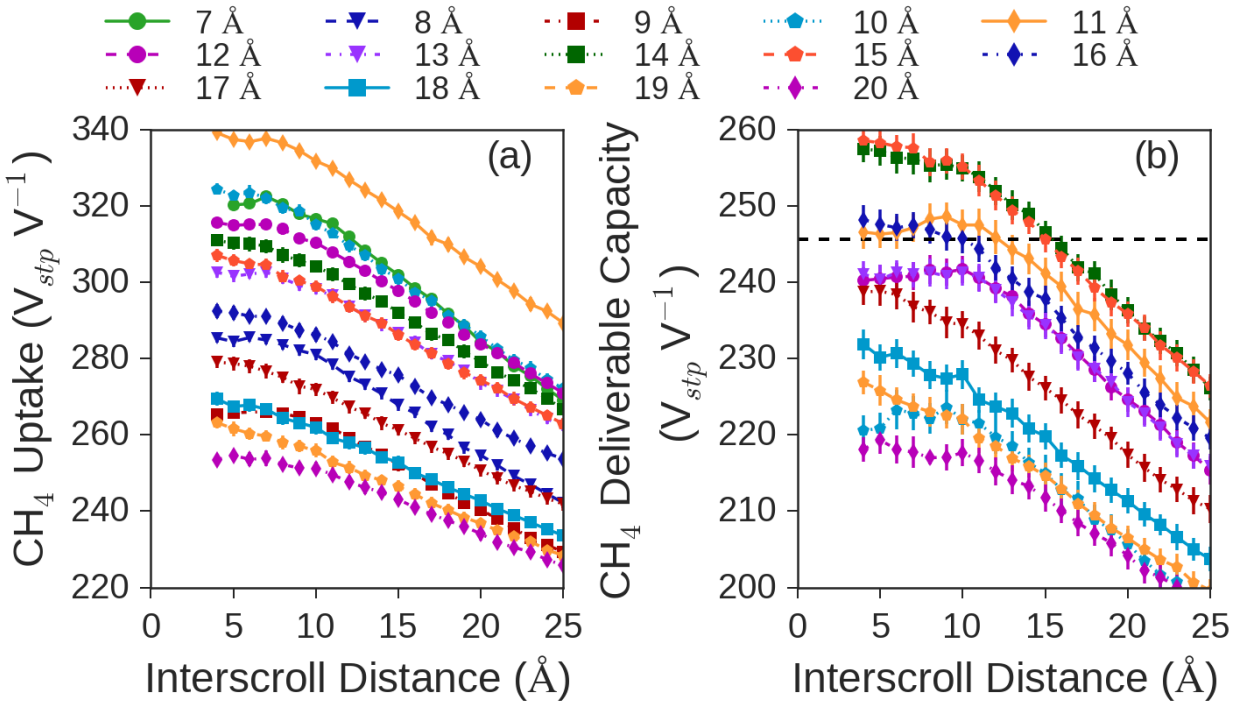

Figure S2. a) Computed CH_4 adsorption capacity and b) deliverable capacity as a function of interscroll distances for various interlayer spacing nanoscrolls. Nanoscrolls with length of 1600 Å were used. The adsorption conditions are at 298 K and 65 bar, while the desorption conditions are 358 K and 5.8 bar. Dashed line in b) indicates 95% of maximum deliverable capacity.