Supporting Information: Density Functional Theory Model for Adsorption-Induced Deformation of Mesoporous Materials with Non-Convex Pore Geometry

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Fluid-fluid parameters of interaction

The coexistence densities, presented on Fig. S1 were calculated by solving the system of equations at different temperatures

$$P_{\text{liquid}} = P_{\text{vapor}} \qquad \mu_{\text{liquid}} = \mu_{\text{vapor}}, \tag{S1}$$

where P and μ are give by Eqs. (10) and (11) from the main text respectively, and compared with experimental data taken from NIST.[1] In order to calculate surface tension of liquid-vapor interface, we used presented cDFT in slit geometry. Then, the surface tension is defined as an excess grand potential

$$\gamma_{\text{lv}} = \frac{\Omega[\rho(\mathbf{r})] + PV}{A}, \tag{S2}$$

where A is the area of liquid-vapor interface, P – pressure in the bulk and V is the system volume. The density profile $\rho(\mathbf{r})$ was obtained in the system without external potential. The length of the system was in the range between 30 d_{HS} and 90 d_{HS}.

The VdW-DFT describes binodal densities (Fig. S1), saturation pressure (Fig. S3) and surface tension (Fig. S2) with acceptable accuracy. Specifically, the saturation pressure is fitted with an error less then 5% at temperatures from 75 K to 85 K. The calculated surface tension departures from experimental data in the range of temperatures 75 K - 110 K less than on 6%. Also, liquid density at 77 K is fitted with the error less than 1%.

Solid-fluid parameters of interaction

Solid-fluid interactions were modeled via Lennard-Jones potential with the parameters taken from Lastoskie et al. $\sigma_{\text{sf}} = 0.3494$ nm and $\epsilon_{\text{ff}}/k_{\text{B}} = 74.23$ K.[2] These parameters provide reasonable agreement with N_2 adsorption on BP 280 carbon.[3] Fig. S4 shows that at low pressure region (below $\simeq 0.3$ relative pressure) the calculated isotherm underestimate and above over-

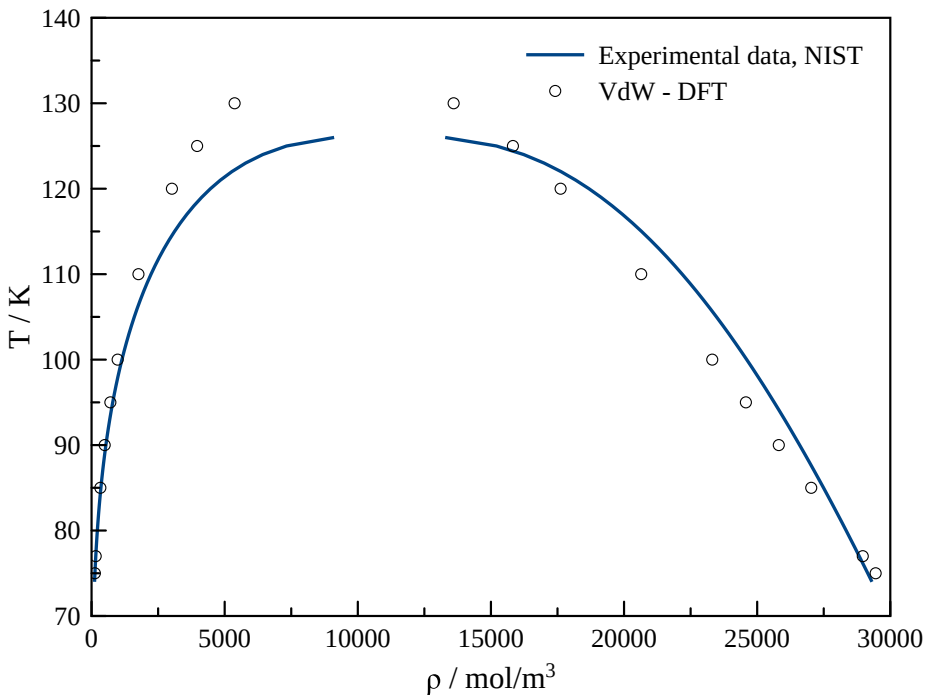

Figure S1: Comparison between experimentally measured binodal of nitrogen and calculations within the present model. Experimental data were taken from NIST.

estimate experimental one. However, the parameters predict the shape and the magnitude of the adsorbed amount on BP 280. This is sufficient for the present study, focusing on general trends of adsorption-induced deformation and not aiming to quantitatively describe neither adsorption nor strain isotherms.

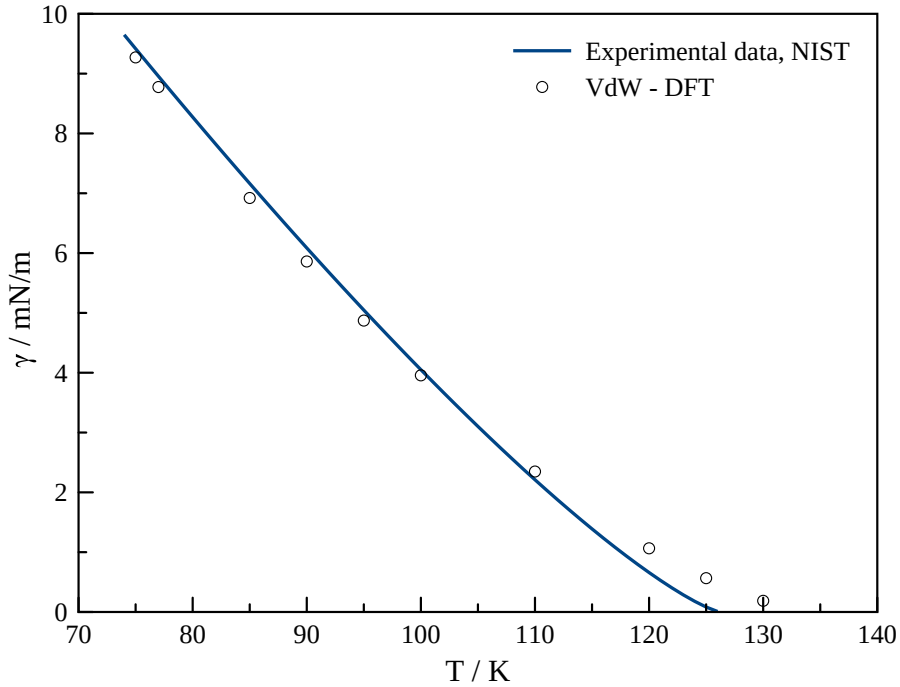

Figure S2: Comparison between experimentally measured surface tension of nitrogen and calculations within the present model. Experimental data were taken from NIST.

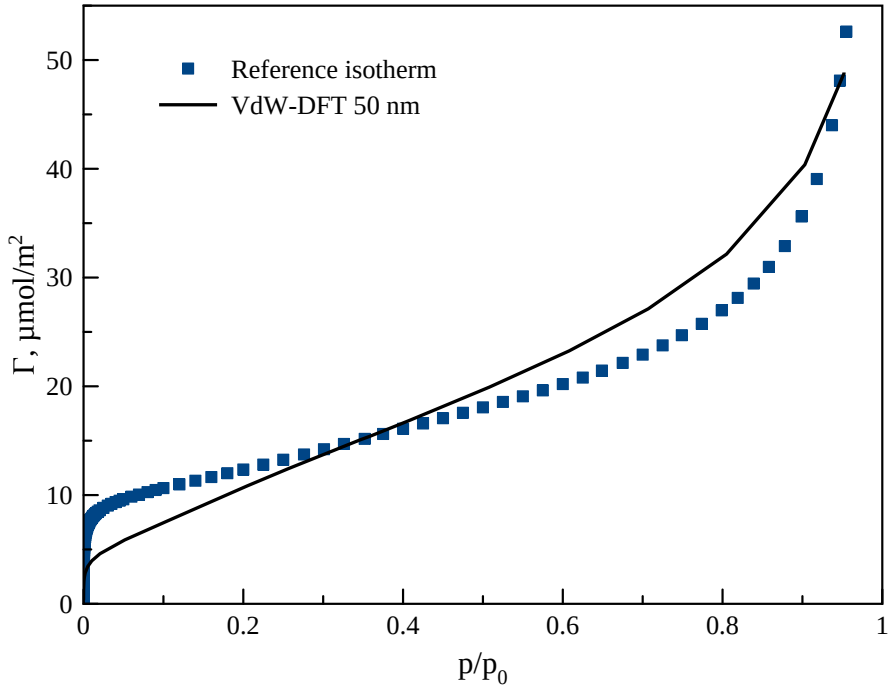

Figure S4: Comparison between standard nitrogen adsorption measured on carbon BP 280[3] and DFT calculations performed in 50 nm slit pore.

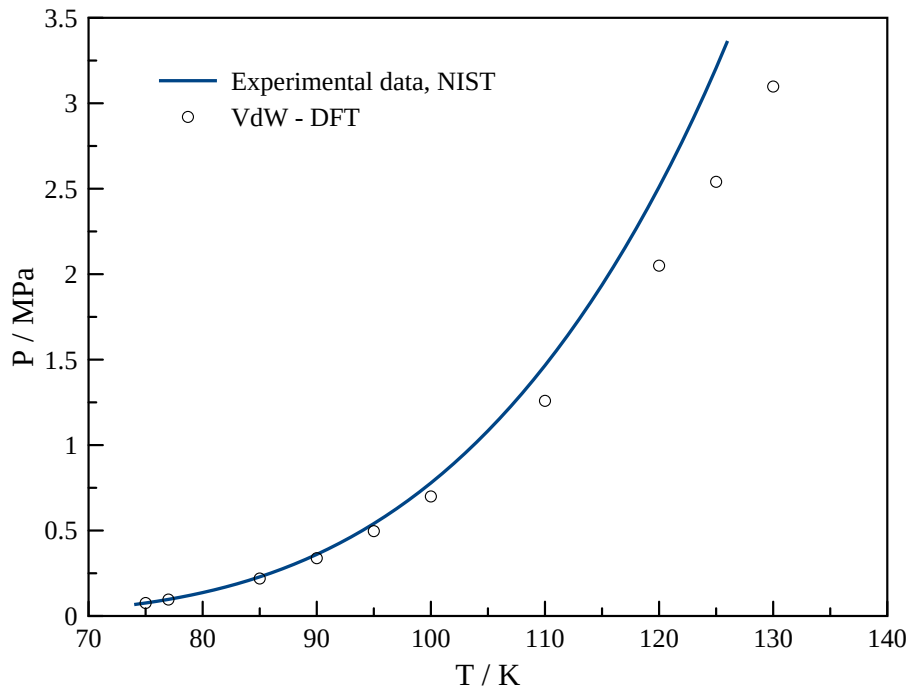

Figure S3: Comparison between experimentally measured vapor pressure of nitrogen and calculations within the present model. Experimental data were taken from NIST.

The calculation was performed in wide slit pore (50 nm) and the excess adsorbed amount of nonporous material we calculated via:

$$\Gamma = \frac{1}{2}\left[\int_0^L dz\rho(z) - \rho_{\mathrm{b}}(L-\sigma_{\mathrm{c}})\right],$$

where σ_{c} is the effective diameter of carbon atom. The DFT was implemented in the slit geometry with the following potential of interactions:

$$V_{\mathrm{ext}}(z) = V_{\mathrm{wall}}(z) + V_{\mathrm{wall}}(L-z), \tag{S3}$$

where V_{wall} is the potential of interactions with the infinite solid surface:

$$V_{\mathrm{wall}}(z) = 8\pi\epsilon_{\mathrm{sf}}\rho_{\mathrm{s}}\sigma_{\mathrm{sf}}^2\left[\frac{1}{10}\left(\frac{\sigma_{\mathrm{sf}}}{z}\right)^{10} - \frac{1}{4}\left(\frac{\sigma_{\mathrm{sf}}}{z}\right)^{4}\right], \tag{S4}$$

also this equation is the limiting form of the external potentials in cylindrical pore and on the external surface of a cylinder.

Tabulated data

Table S1: Cylindrical pore at 77 K

Adsorption			Desorption		
p/p_0	Γ [$\mu mol/m^2$]	f [MPa]	p/p_0	Γ [$\mu mol/m^2$]	f [MPa]
1.030E-10	6.453E-05	-1.681E-05	1.030E-10	6.453E-05	-1.681E-05
1.030E-09	6.443E-04	-1.678E-04	1.030E-09	6.443E-04	-1.678E-04
1.030E-08	6.374E-03	-1.647E-03	1.030E-08	6.374E-03	-1.647E-03
1.030E-07	5.783E-02	-1.391E-02	1.030E-07	5.783E-02	-1.391E-02
1.030E-06	3.374E-01	-4.743E-02	1.030E-06	3.374E-01	-4.743E-02
5.170E-06	7.483E-01	8.767E-03	5.170E-06	7.483E-01	8.767E-03
1.030E-05	9.671E-01	9.074E-02	1.030E-05	9.671E-01	9.074E-02
5.170E-05	1.546E+00	4.750E-01	5.170E-05	1.546E+00	4.750E-01
1.033E-04	1.824E+00	7.373E-01	1.033E-04	1.824E+00	7.373E-01
5.165E-04	2.546E+00	1.607E+00	5.165E-04	2.546E+00	1.607E+00
1.033E-03	2.901E+00	2.105E+00	1.033E-03	2.901E+00	2.105E+00
5.164E-03	3.908E+00	3.595E+00	5.164E-03	3.908E+00	3.595E+00
1.033E-02	4.490E+00	4.397E+00	1.033E-02	4.490E+00	4.397E+00
2.065E-02	5.272E+00	5.307E+00	2.065E-02	5.272E+00	5.307E+00
5.157E-02	7.099E+00	6.664E+00	5.157E-02	7.099E+00	6.664E+00
1.030E-01	9.802E+00	7.844E+00	1.030E-01	9.802E+00	7.844E+00
1.542E-01	1.200E+01	8.729E+00	1.542E-01	1.200E+01	8.729E+00
2.053E-01	1.405E+01	9.408E+00	2.053E-01	1.405E+01	9.408E+00
2.562E-01	1.627E+01	9.806E+00	2.307E-01	1.520E+01	9.607E+00
2.816E-01	1.778E+01	9.692E+00	2.308E-01	2.268E+01	3.445E+00
2.917E-01	2.318E+01	6.846E+00	2.358E-01	2.273E+01	3.758E+00
3.069E-01	2.328E+01	7.599E+00	2.460E-01	2.283E+01	4.362E+00
3.474E-01	2.350E+01	9.453E+00	2.562E-01	2.292E+01	4.947E+00
3.575E-01	2.355E+01	9.885E+00	2.664E-01	2.300E+01	5.511E+00
4.079E-01	2.376E+01	1.189E+01	2.765E-01	2.307E+01	6.058E+00
5.083E-01	2.408E+01	1.529E+01	2.867E-01	2.315E+01	6.587E+00
6.079E-01	2.432E+01	1.809E+01	2.968E-01	2.321E+01	7.101E+00
7.069E-01	2.451E+01	2.048E+01	3.069E-01	2.328E+01	7.599E+00
8.053E-01	2.466E+01	2.255E+01	3.474E-01	2.350E+01	9.453E+00
9.030E-01	2.478E+01	2.438E+01	3.575E-01	2.355E+01	9.885E+00
9.516E-01	2.484E+01	2.522E+01	4.079E-01	2.376E+01	1.189E+01
			5.083E-01	2.408E+01	1.529E+01
			6.079E-01	2.432E+01	1.809E+01
			7.069E-01	2.451E+01	2.048E+01
			8.053E-01	2.466E+01	2.255E+01
			9.030E-01	2.478E+01	2.438E+01
			9.516E-01	2.484E+01	2.522E+01

Table S2: Cylindrical pore at 90 K

Adsorption			Desorption		
p/p_0	Γ [$\mu mol/m^2$]	f [MPa]	p/p_0	Γ [$\mu mol/m^2$]	f [MPa]
1.090E-10	1.494E-05	-2.966E-06	1.090E-10	1.494E-05	-2.966E-06
1.090E-09	1.492E-04	-2.965E-05	1.090E-09	1.492E-04	-2.965E-05
1.090E-08	1.489E-03	-2.950E-04	1.090E-08	1.489E-03	-2.950E-04
1.090E-07	1.455E-02	-2.811E-03	1.090E-07	1.455E-02	-2.811E-03
1.090E-06	1.206E-01	-1.841E-02	1.090E-06	1.206E-01	-1.841E-02
5.450E-06	3.793E-01	-1.973E-02	5.450E-06	3.793E-01	-1.973E-02
1.090E-05	5.535E-01	9.259E-03	1.090E-05	5.535E-01	9.259E-03
5.450E-05	1.077E+00	2.401E-01	5.450E-05	1.077E+00	2.401E-01
1.090E-04	1.344E+00	4.372E-01	1.090E-04	1.344E+00	4.372E-01
5.449E-04	2.055E+00	1.184E+00	5.449E-04	2.055E+00	1.184E+00
1.090E-03	2.409E+00	1.649E+00	1.090E-03	2.409E+00	1.649E+00
5.447E-03	3.410E+00	3.124E+00	5.447E-03	3.410E+00	3.124E+00
1.089E-02	3.982E+00	3.957E+00	1.089E-02	3.982E+00	3.957E+00
2.176E-02	4.730E+00	4.933E+00	2.176E-02	4.730E+00	4.933E+00
5.427E-02	6.332E+00	6.480E+00	5.427E-02	6.332E+00	6.480E+00
1.081E-01	8.523E+00	7.931E+00	1.081E-01	8.523E+00	7.931E+00
1.615E-01	1.035E+01	8.997E+00	1.615E-01	1.035E+01	8.997E+00
2.144E-01	1.199E+01	9.876E+00	2.144E-01	1.199E+01	9.876E+00
2.668E-01	1.358E+01	1.060E+01	2.668E-01	1.358E+01	1.060E+01
3.189E-01	1.529E+01	1.115E+01	3.189E-01	1.530E+01	1.115E+01
3.498E-01	1.662E+01	1.123E+01	3.189E-01	2.050E+01	7.050E+00
3.565E-01	1.710E+01	1.111E+01	3.292E-01	2.062E+01	7.497E+00
3.601E-01	2.091E+01		3.395E-01	2.072E+01	7.936E+00
3.652E-01	2.095E+01	8.998E+00	3.498E-01	2.082E+01	8.368E+00
3.704E-01	2.099E+01	9.204E+00	3.601E-01	2.091E+01	8.790E+00
4.215E-01	2.132E+01	1.114E+01	3.704E-01	2.099E+01	9.204E+00
5.224E-01	2.178E+01	1.447E+01	4.215E-01	2.132E+01	1.114E+01
6.215E-01	2.208E+01	1.722E+01	5.224E-01	2.178E+01	1.447E+01
7.188E-01	2.230E+01	1.956E+01	6.215E-01	2.208E+01	1.722E+01
8.143E-01	2.247E+01	2.158E+01	7.188E-01	2.230E+01	1.956E+01
9.081E-01	2.259E+01	2.335E+01	8.143E-01	2.247E+01	2.158E+01
9.542E-01	2.265E+01	2.415E+01	9.081E-01	2.259E+01	2.335E+01
			9.542E-01	2.265E+01	2.415E+01

Table S3: Cylindrical pore at 110 K

Adsorption		
p/p_0	Γ [$\mu mol/m^2$]	f [MPa]
1.280E-10	3.247E-06	-3.337E-07
1.280E-09	3.245E-05	-3.336E-06
1.280E-08	3.243E-04	-3.331E-05
1.280E-07	3.228E-03	-3.276E-04
1.280E-06	3.089E-02	-2.772E-03
6.400E-06	1.314E-01	-6.182E-03
1.280E-05	2.258E-01	-1.529E-03
6.390E-05	6.095E-01	9.585E-02
1.279E-04	8.424E-01	2.153E-01
6.394E-04	1.517E+00	7.943E-01
1.279E-03	1.866E+00	1.208E+00
6.388E-03	2.872E+00	2.657E+00
1.276E-02	3.448E+00	3.536E+00
2.547E-02	4.193E+00	4.610E+00
6.323E-02	5.703E+00	6.420E+00
1.250E-01	7.598E+00	8.227E+00
1.854E-01	9.142E+00	9.562E+00
2.444E-01	1.049E+01	1.067E+01
3.019E-01	1.175E+01	1.162E+01
3.581E-01	1.299E+01	1.243E+01
4.020E-01	1.406E+01	1.295E+01
4.128E-01	1.437E+01	1.304E+01
4.343E-01	1.539E+01	1.296E+01
4.370E-01	1.678E+01	1.198E+01
4.662E-01	1.736E+01	1.260E+01
5.687E-01	1.814E+01	1.525E+01
6.657E-01	1.850E+01	1.754E+01
7.573E-01	1.869E+01	1.945E+01
8.435E-01	1.879E+01	2.105E+01
9.244E-01	1.882E+01	2.241E+01
9.629E-01	1.883E+01	2.300E+01

Table S4: CMK-3 at 77 K

Adsorption			Desorption		
p/p_0	Γ [$\mu mol/m^2$]	f [MPa]	p/p_0	Γ [$\mu mol/m^2$]	f [MPa]
1.033E-03	2.248E+00	-3.729E-02	1.033E-03	2.248E+00	-3.729E-02
2.066E-03	2.652E+00	-4.569E-02	2.066E-03	2.652E+00	-4.569E-02
5.164E-03	3.257E+00	-5.838E-02	5.164E-03	3.257E+00	-5.838E-02
1.033E-02	3.794E+00	-6.897E-02	1.033E-02	3.794E+00	-6.897E-02
2.065E-02	4.439E+00	-8.087E-02	2.065E-02	4.439E+00	-8.087E-02
5.157E-02	5.609E+00	-1.020E-01	5.157E-02	5.609E+00	-1.020E-01
1.030E-01	7.048E+00	-1.316E-01	1.030E-01	7.048E+00	-1.316E-01
1.542E-01	8.434E+00	-1.655E-01	1.542E-01	8.434E+00	-1.655E-01
2.053E-01	9.900E+00	-2.074E-01	2.053E-01	9.900E+00	-2.074E-01
2.562E-01	1.143E+01	-2.589E-01	2.562E-01	1.143E+01	-2.589E-01
2.867E-01	1.237E+01	-2.961E-01	2.867E-01	1.237E+01	-2.961E-01
3.069E-01	1.299E+01	-3.250E-01	3.069E-01	1.299E+01	-3.250E-01
3.272E-01	1.362E+01	-3.593E-01	3.230E-01	1.330E+01	-3.421E-01
3.575E-01	1.456E+01	-4.297E-01	3.230E-01	2.307E+01	-9.693E+00
3.878E-01	1.555E+01	-5.546E-01	3.272E-01	2.398E+01	-9.989E+00
3.979E-01	1.590E+01	-6.272E-01	3.575E-01	2.652E+01	-1.061E+01
4.029E-01	1.609E+01	-6.770E-01	3.710E-01	2.776E+01	-1.084E+01
4.079E-01	1.628E+01		3.710E-01	4.186E+01	-1.684E+01
4.180E-01	1.670E+01		3.878E-01	4.203E+01	-1.618E+01
4.280E-01	3.285E+01	-1.178E+01	3.979E-01	4.213E+01	-1.575E+01
4.331E-01	3.354E+01		4.029E-01	4.217E+01	-1.554E+01
4.381E-01	3.453E+01		4.079E-01	4.222E+01	-1.534E+01
4.431E-01	4.253E+01	-1.396E+01	4.180E-01	4.231E+01	-1.493E+01
4.481E-01	4.257E+01	-1.377E+01	4.280E-01	4.240E+01	-1.454E+01
4.532E-01	4.260E+01	-1.358E+01	4.331E-01	4.244E+01	-1.434E+01
4.582E-01	4.264E+01	-1.339E+01	4.381E-01	4.248E+01	-1.415E+01
4.782E-01	4.279E+01	-1.267E+01	4.431E-01	4.253E+01	-1.396E+01
5.083E-01	4.299E+01	-1.163E+01	4.481E-01	4.257E+01	-1.377E+01
5.582E-01	4.329E+01	-1.002E+01	4.532E-01	4.260E+01	-1.358E+01
6.079E-01	4.355E+01	-8.545E+00	4.582E-01	4.264E+01	-1.339E+01
7.069E-01	4.398E+01	-5.904E+00	4.782E-01	4.279E+01	-1.267E+01
8.053E-01	4.432E+01	-3.600E+00	5.083E-01	4.299E+01	-1.163E+01
9.030E-01	4.460E+01	-1.559E+00	5.582E-01	4.329E+01	-1.002E+01
			6.079E-01	4.355E+01	-8.545E+00
			7.069E-01	4.398E+01	-5.904E+00

Table S5: CMK-3 at 90 K

Adsorption			Desorption		
p/p_0	Γ [$\mu mol/m^2$]	f [MPa]	p/p_0	Γ [$\mu mol/m^2$]	f [MPa]
1.090E-03	1.809E+00	-2.682E-02	1.090E-03	1.809E+00	-2.682E-02
2.180E-03	2.213E+00	-3.265E-02	2.180E-03	2.213E+00	-3.265E-02
5.447E-03	2.828E+00	-4.172E-02	5.447E-03	2.828E+00	-4.172E-02
1.089E-02	3.379E+00	-4.923E-02	1.089E-02	3.379E+00	-4.923E-02
2.176E-02	4.050E+00	-5.700E-02	2.176E-02	4.050E+00	-5.700E-02
5.427E-02	5.283E+00	-6.909E-02	5.427E-02	5.283E+00	-6.909E-02
1.081E-01	6.803E+00	-8.547E-02	1.081E-01	6.803E+00	-8.547E-02
1.615E-01	8.215E+00	-1.055E-01	1.615E-01	8.215E+00	-1.055E-01
2.144E-01	9.632E+00	-1.321E-01	2.144E-01	9.632E+00	-1.321E-01
2.668E-01	1.106E+01	-1.691E-01	2.668E-01	1.106E+01	-1.691E-01
2.981E-01	1.193E+01	-2.008E-01	2.981E-01	1.193E+01	-2.008E-01
3.189E-01	1.252E+01	-2.292E-01	3.189E-01	1.252E+01	-2.292E-01
3.395E-01	1.311E+01	-2.672E-01	3.395E-01	1.311E+01	-2.672E-01
3.704E-01	1.405E+01	-3.577E-01	3.704E-01	1.405E+01	-3.577E-01
4.011E-01	1.515E+01	-5.526E-01	3.704E-01	2.070E+01	-6.568E+00
4.113E-01	2.367E+01		4.011E-01	2.298E+01	-7.431E+00
4.164E-01	2.401E+01		4.113E-01	2.368E+01	-7.625E+00
4.215E-01	2.435E+01		4.164E-01	2.403E+01	-7.711E+00
4.317E-01	2.503E+01	-7.934E+00	4.215E-01	2.437E+01	-7.791E+00
4.419E-01	2.571E+01	-8.060E+00	4.317E-01	2.505E+01	-7.934E+00
4.520E-01	2.638E+01	-8.170E+00	4.419E-01	2.572E+01	-8.060E+00
4.621E-01	2.706E+01	-8.269E+00	4.520E-01	2.640E+01	-8.170E+00
4.722E-01	2.776E+01	-8.360E+00	4.621E-01	2.706E+01	-8.269E+00
4.923E-01	2.924E+01	-8.536E+00	4.621E-01	3.745E+01	-1.281E+01
5.224E-01	3.813E+01		4.722E-01	3.758E+01	-1.247E+01
6.215E-01	3.893E+01	-7.908E+00	4.923E-01	3.782E+01	-1.180E+01
7.188E-01	3.949E+01	-5.394E+00	5.224E-01	3.813E+01	-1.083E+01
8.143E-01	3.990E+01	-3.203E+00	6.215E-01	3.893E+01	-7.908E+00
9.081E-01	4.023E+01	-1.271E+00	7.188E-01	3.949E+01	-5.394E+00
			8.143E-01	3.990E+01	-3.203E+00

Table S6: CMK-3 at 110 K

Adsorption			Desorption		
p/p_0	Γ [$\mu mol/m^2$]	f [MPa]	p/p_0	Γ [$\mu mol/m^2$]	f [MPa]
1.279E-03	1.346E+00	-1.908E-02	1.279E-03	1.346E+00	-1.908E-02
2.557E-03	1.744E+00	-2.358E-02	2.557E-03	1.744E+00	-2.358E-02
6.388E-03	2.369E+00	-2.900E-02	6.388E-03	2.369E+00	-2.900E-02
1.276E-02	2.944E+00	-3.085E-02	1.276E-02	2.944E+00	-3.085E-02
2.547E-02	3.660E+00	-2.761E-02	2.547E-02	3.660E+00	-2.761E-02
6.323E-02	5.010E+00	-7.166E-03	6.323E-02	5.010E+00	-7.166E-03
1.250E-01	6.681E+00	2.881E-02	1.250E-01	6.681E+00	2.881E-02
1.854E-01	8.180E+00	5.695E-02	1.854E-01	8.180E+00	5.695E-02
2.444E-01	9.624E+00	6.772E-02	2.444E-01	9.624E+00	6.772E-02
3.019E-01	1.106E+01	3.674E-02	3.019E-01	1.106E+01	3.674E-02
3.358E-01	1.196E+01	-2.860E-02	3.358E-01	1.196E+01	-2.860E-02
3.581E-01	1.260E+01	-1.133E-01	3.581E-01	1.260E+01	-1.133E-01
3.691E-01	1.295E+01	-1.777E-01	3.691E-01	1.295E+01	-1.777E-01
3.801E-01	1.333E+01	-2.658E-01	3.801E-01	1.333E+01	-2.658E-01
3.911E-01	1.376E+01	-3.929E-01	3.911E-01	1.376E+01	-3.929E-01
4.020E-01	1.434E+01	-5.979E-01	4.020E-01	1.434E+01	-5.979E-01
4.128E-01	1.582E+01	-1.097E+00	4.128E-01	1.582E+01	-1.097E+00
4.236E-01	1.706E+01	-2.358E+00	4.236E-01	1.706E+01	-2.358E+00
4.343E-01	1.786E+01	-3.125E+00	4.343E-01	1.786E+01	-3.125E+00
4.450E-01	1.853E+01	-3.493E+00	4.450E-01	1.853E+01	-3.493E+00
4.662E-01	1.973E+01	-3.947E+00	4.662E-01	1.973E+01	-3.947E+00
5.181E-01	2.235E+01	-4.487E+00	5.181E-01	2.235E+01	-4.487E+00
5.687E-01	2.485E+01	-4.688E+00	5.687E-01	2.485E+01	-4.688E+00
5.885E-01	2.601E+01	-4.759E+00	5.885E-01	2.601E+01	-4.759E+00
5.984E-01	2.682E+01	-4.838E+00	5.885E-01	3.026E+01	-6.423E+00
6.033E-01	3.054E+01		5.984E-01	3.046E+01	-6.230E+00
6.082E-01	3.063E+01		6.033E-01	3.055E+01	-6.131E+00
6.657E-01	3.137E+01	-4.828E+00	6.082E-01	3.063E+01	-6.032E+00
7.573E-01	3.208E+01	-2.969E+00	6.657E-01	3.137E+01	-4.828E+00
8.435E-01	3.248E+01	-1.353E+00	7.573E-01	3.208E+01	-2.969E+00
9.244E-01	3.270E+01	3.627E-02	8.435E-01	3.248E+01	-1.353E+00

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