Supporting Information

Impact of mechanical deformation on guest diffusion in zeolitic imidazolate frameworks

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Table S1. Force Field Parameters for ZIF-8 Flexible Framework.\textsuperscript{1}

\begin{table}[h]
\centering
\begin{tabular}{lll}
\hline
 & bond potential: $E_{\text{bond}} = K_\theta (\theta - \theta_0)^2$ & \\
 & bond type & $K_\theta$ (kcal mol\textsuperscript{-1} Å\textsuperscript{-2}) & $\theta_0$ (Å) \\
\hline
C1-C3 & 346.543 & 1.490 \hline
C1-N & 488.000 & 1.335 \hline
C2-N & 440.210 & 1.370 \hline
C2-H2 & 367.000 & 1.080 \hline
C2-C2 & 540.249 & 1.350 \hline
C3-H3 & 340.000 & 1.090 \hline
Zn-N & 78.500 & 2.011 \hline
\end{tabular}
\end{table}

\begin{table}[h]
\centering
\begin{tabular}{llllll}
\hline
 & dihedral: $E_{\text{proper}} = K_\phi [1 + \cos(n(\varphi - \varphi_0))]$ & \\
 & type\textsuperscript{a} & $K_\phi$ (kcal mol\textsuperscript{-1]) & n & $\varphi_0$ (°) \\
\hline
X-N-C2-X & 2.325 & 2 & 180.0 \hline
X-C2-C2-X & 5.150 & 2 & 180.0 \hline
X-C1-N-X & 5.000 & 2 & 180.0 \hline
\end{tabular}
\end{table}

\begin{table}[h]
\centering
\begin{tabular}{llll}
\hline
 & improper: $E_{\text{improper}} = K_\phi (\varphi - \varphi_0)^2$ & \\
 & type\textsuperscript{b} & $K_\phi$ (kcal mol\textsuperscript{-1} rad\textsuperscript{-2}) & $\varphi_0$ (°) \\
\hline
N-C3-C1-N & 2.000 & 180.0 \hline
C2-H2-C2-N & 2.000 & 180.0 \hline
C2-Zn-N-C1 & 2.000 & 180.0 \hline
\end{tabular}
\end{table}

\begin{table}[h]
\centering
\begin{tabular}{llll}
\hline
 & VDW interaction & partial charges & \\
 & atom & $\epsilon$ (kcal/mol) & $\sigma$ (Å) & $q$ (e) \\
\hline
Zn & 0.0125 & 1.960 & +0.7362 \hline
N & 0.1700 & 3.250 & -0.3008 \hline
C1 & 0.0860 & 3.400 & +0.4339 \hline
C2 & 0.0860 & 3.400 & -0.1924 \hline
C3 & 0.1094 & 3.400 & -0.6024 \hline
H2 & 0.0150 & 2.511 & +0.1585 \hline
H3 & 0.0157 & 2.650 & +0.1572 \hline
\end{tabular}
\end{table}

\textsuperscript{a} X denotes wildcard atoms: the given potential term is repeated for each possible dihedral combination X-A-B-X having A-B as central atoms.

\textsuperscript{b} Improper term A-B-C-D refers to the angle between planes A-B-C and B-C-D, with B-C axis of rotation.
**Table S2.** Force Field Parameters for CO$_2$ Molecules$^{82}$ and H$_2$ Molecules$^{83}$.

<table>
<thead>
<tr>
<th>atom type</th>
<th>$\varepsilon$ (kcal/mol)</th>
<th>$\sigma$ (Å)</th>
<th>$q$ (e)</th>
</tr>
</thead>
<tbody>
<tr>
<td>C</td>
<td>0.05584</td>
<td>2.757</td>
<td>+ 0.6512</td>
</tr>
<tr>
<td>O</td>
<td>0.15982</td>
<td>3.033</td>
<td>− 0.3256</td>
</tr>
<tr>
<td>H</td>
<td>0.00147</td>
<td>3.314</td>
<td>0.0000</td>
</tr>
</tbody>
</table>

**Figure S1.** Linear fitting the stress-strain data to obtain (a) the Young’s modulus and (b) the shear modulus of ZIF-8.
H2ZIF8.indata: including the initial configuration of H2ZIF8 system and all related force field parameters.

CO2ZIF8.indata: including the initial configuration of CO2ZIF8 system and all related force field parameters.

CO2ZIF8(NVE)-unstrained.inrun: using CO2ZIF8.indata as input to perform NVE equilibrium the system. It will output the file “0.restart.50000000”.

CO2ZIF8(NVE)-tenisle.inrun: using “0.restart.50000000” as starting point to perform uniaxial tensile deformation of ZIF-8. At every strain point, the system will be performed NVE equilibrium for 50 ns to collect data for guest diffusion computation.

CO2ZIF8(NVE)-shear.inrun: using “0.restart.50000000” as starting point to perform shear deformation of ZIF-8. At every strain point, the system will be performed NVE equilibrium for 50 ns to collect data for guest diffusion computation.

CO2ZIF8(NPT)-unstrained.inrun: using CO2ZIF8.indata as input to perform NPT equilibrium the system. It will output the file “0.restart.50000000”. It should be noted that the different commands were used for later tensile and shear deformation (marked inside this file).

CO2ZIF8(NPT)-tenisle.inrun: using “0.restart.50000000” as starting point to perform uniaxial tensile deformation of ZIF-8. At every strain point, the system will be performed NPT equilibrium for 50 ns to collect data for guest diffusion computation.

CO2ZIF8(NPT)-shear.inrun: using “0.restart.50000000” as starting point to perform shear deformation of ZIF-8. At every strain point, the system will be performed NPT equilibrium for 50 ns to collect data for guest diffusion computation.

Regarding to H2ZIF8 system, you only have to replace the character of “CO2” with the “H2” in above files with suffix “inrun”.

Calculate the self-diffusion rate by mean squared displacement using the Einstein relation:

\[
D_S = \frac{1}{2d_0} \lim_{t \to \infty} \frac{d}{dt} \left\langle \frac{1}{N} \sum_{i=1}^{N} [\mathbf{r}_i(t) - \mathbf{r}_i(t_0)]^2 \right\rangle
\]

where \(d_0\) is the dimensionality of the system and \(\mathbf{r}_i(t)\) is the position vector of the sorbate molecule \(i\) at time \(t\). The \(D_S\) value is averaged over all the \(N\) sorbed molecules and over multiple time origins \(t_0\) (as symbolized by the brackets).

References