I. Force fields

A. The CO$_2$ Model

The form of the potential of CO$_2$/ε$^3$ and the CO$_2$/TraPPEFlex$^4$ models, consist of intra and intermolecular potentials. In fact, for a flexible model an harmonic potentials was included in the intramolecular interaction.

\[ U(\theta) = \frac{k_\theta}{2} (\theta - \theta_0)^2, \quad (S.1) \]

where $\theta$ is the angle O-C-O and $\theta_0$ refers to the equilibrium value, $k_\theta$ is the spring constant. For the intermolecular potentail, between two CO$_2$ molecules, the LJ and Coulomb interactions are used,

\[ u(r) = 4\epsilon_{\alpha\beta} \left[ \left( \frac{\sigma_{\alpha\beta}}{r} \right)^{12} - \left( \frac{\sigma_{\alpha\beta}}{r} \right)^6 \right] + \frac{1}{4\pi\epsilon_0} \frac{q_\alpha q_\beta}{r} \quad (S.2) \]
TABLE S 1: Parameters of the CO$_2$ models considered in this work.

<table>
<thead>
<tr>
<th>model</th>
<th>$d_{OC}$</th>
<th>$k_{\theta}$</th>
<th>$\theta_{OCO}$</th>
<th>$\epsilon_{O-O}$</th>
<th>$\sigma_{O-O}$</th>
<th>$\epsilon_{C-C}$</th>
<th>$\sigma_{C-C}$</th>
<th>$q_{O}$</th>
<th>$q_{C}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>CO$_2$/\epsilon</td>
<td>1.170</td>
<td>500</td>
<td>180</td>
<td>0.1484</td>
<td>2.363</td>
<td>0.9229</td>
<td>2.62585</td>
<td>-0.6204</td>
<td>1.2408</td>
</tr>
<tr>
<td>CO$_2$/TraPPE</td>
<td>1.160</td>
<td>1236</td>
<td>180</td>
<td>2.7481</td>
<td>3.050</td>
<td>0.9392</td>
<td>2.8</td>
<td>-0.350</td>
<td>0.70</td>
</tr>
</tbody>
</table>

where $r$ is the distance between sites $\alpha$ and $\beta$, $q_\alpha$ is the electric charge of site $\alpha$, $\epsilon_0$ is the permitivity of vacuum, $\epsilon_{\alpha\beta}$ is the LJ energy scale and $\sigma_{\alpha\beta}$ the repulsive diameter for an $\alpha-\beta$ pair. The cross interactions between unlike atoms are obtained using the Lorentz-Berthelot mixing rules,

$$\sigma_{\alpha\beta} = \left(\frac{\sigma_{\alpha\alpha} + \sigma_{\beta\beta}}{2}\right); \quad \epsilon_{\alpha\beta} = \left(\epsilon_{\alpha\alpha}\epsilon_{\beta\beta}\right)^{1/2}$$  \hspace{1cm} (S.3)

B. Grafene.

The force field of grafene is model with fixed carbon atoms, Lennard-Jones interaction and a uniform surface charge densities, $\sigma_S$ in each layer with opposite sign.

TABLE S 2: Force field parameters of grafene model.

<table>
<thead>
<tr>
<th>Model</th>
<th>$q_C/e$</th>
<th>$\sigma/\rm{Å}$</th>
<th>$(\epsilon/k_B)/\rm{K}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>C</td>
<td>0.0143$^\dagger$</td>
<td>3.401</td>
<td>35.46</td>
</tr>
</tbody>
</table>
II. Statistical calculation

A. Dielectric constant, $\epsilon$

It is obtained from the fluctuations of the system’s total dipole moment $M = \sum_i q_i r_i$,

$$\epsilon = 1 + \frac{4\pi}{3k_B TV} (\langle M^2 \rangle - \langle M \rangle^2)$$  \hspace{1cm} (S.4)

where $r_i$ is the position of atom $i$, $k_B$ is the Boltzmann constant, $T$ the absolute temperature and $V$ the simulation cell volume.

B. The self-diffusion coefficient, $D$

It is obtained from the Einstein equation

$$D = \lim_{t \to \infty} \frac{1}{6t} \langle |\mathbf{R}_i(t) - \mathbf{R}_i(0)|^2 \rangle,$$  \hspace{1cm} (S.5)

where $\mathbf{R}_i(t)$ is the center of mass position of molecule $i$ at time $t$ and $\langle ... \rangle$ denotes time average.

C. Stored energy.

Since the capacitance is related with the energy storage in the capacitor; the energy in the systems can be calculated with the electrostatic relation,

$$U = \frac{1}{2} CV^2$$  \hspace{1cm} (S.6)
Figure 1: Electrostatic energy, in log scale, as function of the electric potential for the two CO$_2$ models.

References


