A DFT-D study on the reaction mechanism of selective catalytic reduction of NO by NH₃ over the Fe₂O₃/Ni (111) surface

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Theoretical methods

All DFT calculations were carried out in Vienna Ab-initio Simulation Package (VASP) version 5.4 [1-3]. The generalized gradient approximation (GGA) functional and Perdew-Burke-Ernzerhof (PBE) exchange association functional were used in the DFT calculation [4-6]. The Kohn-Sham equations were solved by the Projector Augmented Wave (PAW) method [7]. The reaction routes of gas molecules on the Fe₂O₃/Ni catalyst surface were investigated by the climbing-image nudged elastic band (CI-NEB) method [8-10]. To further understand the Van der Waals interaction between Ni surface and cluster or gas molecules, we introduced the density functional theory based on dispersion correction (DFT-D3) [11, 12]. In addition, for the convergence criteria of calculation system, the change of total energy and force acting on each ion should be less 10⁻⁵ eV and 10⁻³ eV/Å, respectively.

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Figure S1 Convergence tests of Ni bulk model.

Table S1 Surface energies of Ni slab model.

<table>
<thead>
<tr>
<th>Slab surface</th>
<th>Ni (111)</th>
<th>Ni (100)</th>
<th>Ni (110)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Surface energy (eV)</td>
<td>23.36</td>
<td>27.25</td>
<td>38.94</td>
</tr>
</tbody>
</table>
**Table S2** Double-molecule adsorption energies and charge changes over the Fe$_2$O$_3$/Ni (111) surface.

<table>
<thead>
<tr>
<th>Gas</th>
<th>Adsorption energy (eV)</th>
<th>Bader charge changes (e)</th>
</tr>
</thead>
<tbody>
<tr>
<td>NH$_3$ + NO</td>
<td>-2.935</td>
<td>NH$_3$: -0.141, NO: +0.280</td>
</tr>
<tr>
<td>O$_2$ + NH$_3$</td>
<td>-3.005</td>
<td>O$_2$: +0.455, NH$_3$: -0.132</td>
</tr>
<tr>
<td>O$_2$ + NO</td>
<td>-4.546</td>
<td>O$_2$: +0.380, NO: +0.268</td>
</tr>
</tbody>
</table>

**Figure S2** PDOS results of double-molecule adsorption on the Fe$_2$O$_3$/Ni (111) surface.
**Figure S3** Dissociation path of NHNO\(^*\)(Ni: dark blue, Fe: gold, O: red, N: blue, H: white).

**Figure S4** Dissociation path of NHNO\(_2\)\(^*\)(Ni: dark blue, Fe: gold, O: red, N: blue, H: white).
References


