Supporting Information

First-Principles Study of the Reaction Mechanism of CHO + H on Graphene Surface

Megumi Kayanuma,*,1,2 Mitsuo Shoji,2 Kenji Furuya,2 Katsumasa Kamiya,3 Yuri Aikawa,4
Masayuki Umemura,2 Yasuteru Shigeta2,5

1 Research Center for Computational Design of Advanced Functional Materials, National Institute of Advanced Industrial Science and Technology, Central 2, 1-1-1 Umezono, Tsukuba, Ibaraki 305-8568, Japan
2 Center for Computational Sciences, University of Tsukuba, 1-1-1 Tennodai, Tsukuba, Ibaraki 305-8577, Japan
3 Center for Basic Education and Integrated Learning, Kanagawa Institute of Technology, 1030, Shimoogino, Atsugi, Kanagawa 243-0292, Japan
4 Department of Astronomy, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-0033, Japan
5 Institute of Space and Astronautical Science, Japan Aerospace Exploration Agency, 3-1-1 Yoshinodai, Chuo-ku, Sagamihara, Kanagawa, 252-0222, Japan

Contents

Fig. S1. Initial positions of the free H S2
Fig. S2. Schematic illustration of potential energy curves of molecules adsorbed on surface S2
Fig. S3. A structure after MD simulation in Case 1, which generated CO + H2 S2
Fig. S4. CHOH generated in Case 1 and Case 2 S3
Fig. S5. Optimized structures and transition states on a model graphene surface S3
Fig. S6. Optimized structures of OH on a graphene model and a water molecule S4
Fig. S7. Schematic illustration of reaction paths in Case 1 and Case 2 S4
Table S1. Relative energies of transition states and products of reaction of CHOH on a graphene model S4
**Figure S1.** Initial positions of the free H for (a) Case 1 and (2) Case 2.

**Figure S2.** Schematic illustration of the potential energy curves of the molecules which can (black solid line) and cannot (red dashed line) chemisorb on the surface. When the molecule changes the nature upon reaction, i.e., the reactant (CHO) can chemisorb while the product (CO) cannot, the repulsive force is generated.

**Figure S3.** Top and side views of a structure after MD simulation in Case 1 \(((x,y,z) = (0,0,6))\), which generated CO + H₂.
Figure S4. CHOH generated in some trajectories in (a) Case 1 and (b) Case 2.

Figure S5. Optimized structures of (a) chemisorbed CHOH and the transition state structures of (b) CHOH → CH₂O, (c) desorption of CHOH, (d) CHOH → CHO + H (chemisorbed at ortho position), (e) CHOH → CHO + H (chemisorbed at meta position), (f) CHOH → CHO + H (chemisorbed at para position), (g) CHO + H (chemisorbed at ortho position) → CH₂O, and (h) CHO + H (chemisorbed at para position) → CH₂O on the graphene model.
Figure S6. Optimized structures of OH on (a) a graphene model ($C_{54}H_{18}$) and (b) a water molecule.

Figure S7. Schematic illustrations of reaction paths in (a) Case 1 and (b) Case 2.

Table S1. Relative energies (kJ/mol) of the transition states ($E^*$) and products ($E$) of the reactions on the graphene model ($C_{54}H_{18}$) relative to the energy of CHOH chemisorbed on $C_{54}H_{18}$.

<table>
<thead>
<tr>
<th>reactant</th>
<th>product</th>
<th>$E^*$</th>
<th>$E$</th>
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<tbody>
<tr>
<td>CHO (dissociated)</td>
<td>CH$_2$O</td>
<td>291.6</td>
<td>-196.9</td>
</tr>
<tr>
<td>CHO + H (chemisorbed, ortho)</td>
<td>CH$_2$O</td>
<td>77.3</td>
<td>18.6</td>
</tr>
<tr>
<td>CHO + H (chemisorbed, meta)</td>
<td>99.6</td>
<td>-5.6</td>
<td></td>
</tr>
<tr>
<td>CHO + H (chemisorbed, para)</td>
<td>161.5</td>
<td>155.4</td>
<td></td>
</tr>
<tr>
<td>CHO + H (chemisorbed, ortho)</td>
<td>116.5</td>
<td>1.6</td>
<td></td>
</tr>
<tr>
<td>CHO + H (chemisorbed, para)</td>
<td>153.4</td>
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<td></td>
</tr>
<tr>
<td>CHO + H (chemisorbed, ortho)</td>
<td>CH$_2$O</td>
<td>127.5</td>
<td>-196.9</td>
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