

Supporting Information

**Large-Scale Molecular Dynamics Simulation for  
Ground and Excited States Based on  
Divide-and-Conquer Long-Range Corrected  
Density-Functional Tight-Binding Method**

Nana Komoto<sup>†</sup>, Takeshi Yoshikawa<sup>‡</sup>, Yoshifumi Nishimura<sup>‡</sup>, Hiromi Nakai<sup>\*,†,‡,§</sup>

<sup>†</sup> *Department of Chemistry and Biochemistry, School of Advanced Science and Engineering, Waseda University, 3-4-1 Okubo, Shinjuku-ku, Tokyo 169-8555, Japan*

<sup>‡</sup> *Waseda Research Institute for Science and Engineering, Waseda University, 3-4-1 Okubo, Shinjuku-ku, Tokyo 169-8555, Japan*

<sup>§</sup> *Elements Strategy Initiative for Catalysts and Batteries (ESICB), Kyoto University, Katsura, Kyoto 615-8520, Japan*

---

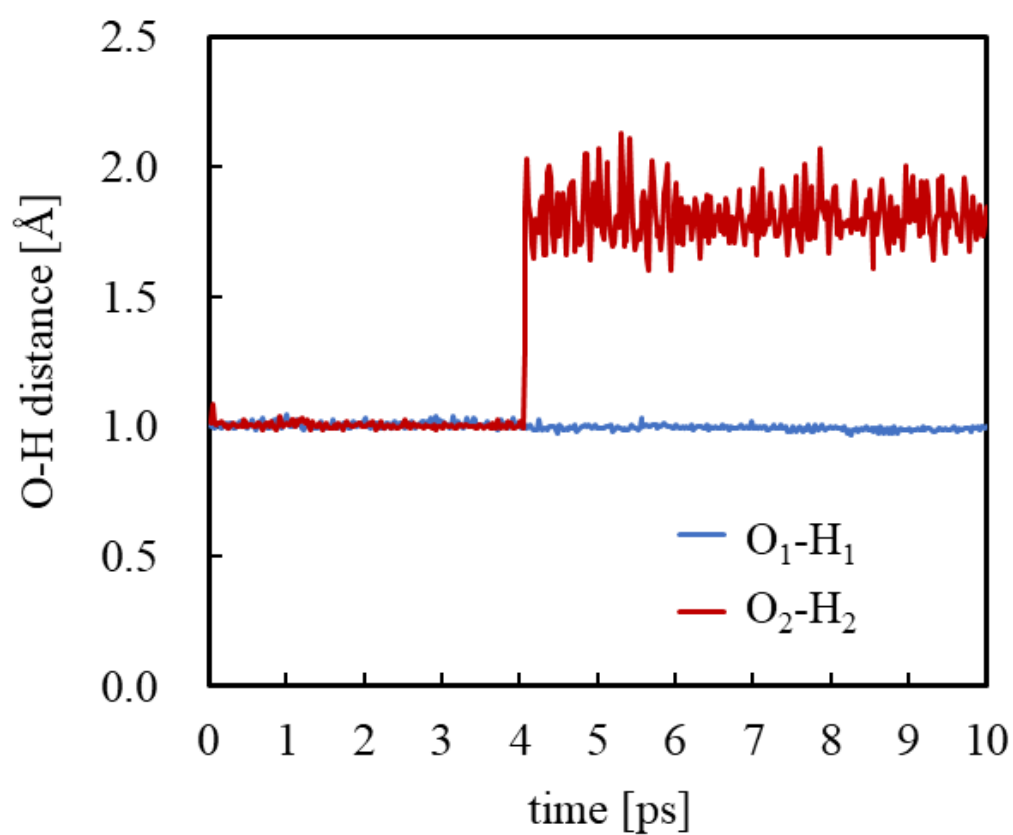
\* Corresponding author. FAX: +81-3-3205-2504,

*E-mail address:* nakai@waseda.jp (H. Nakai).

*URL:* <http://www.chem.waseda.ac.jp/nakai/> (H. Nakai).

## Excited-state molecular dynamics simulation for BP(OH)<sub>2</sub> in gas phase

Figure S1 shows the time-course changes for BP(OH)<sub>2</sub> of the distances,  $r(\text{O}_1\text{-H}_1)$ , between O<sub>1</sub> and H<sub>1</sub> and  $r(\text{O}_2\text{-H}_2)$  between O<sub>2</sub> and H<sub>2</sub>. In the excited state for BP(OH)<sub>2</sub> in gas phase,  $r(\text{O}_2\text{-H}_2)$  greatly increased once at ~4 ps.

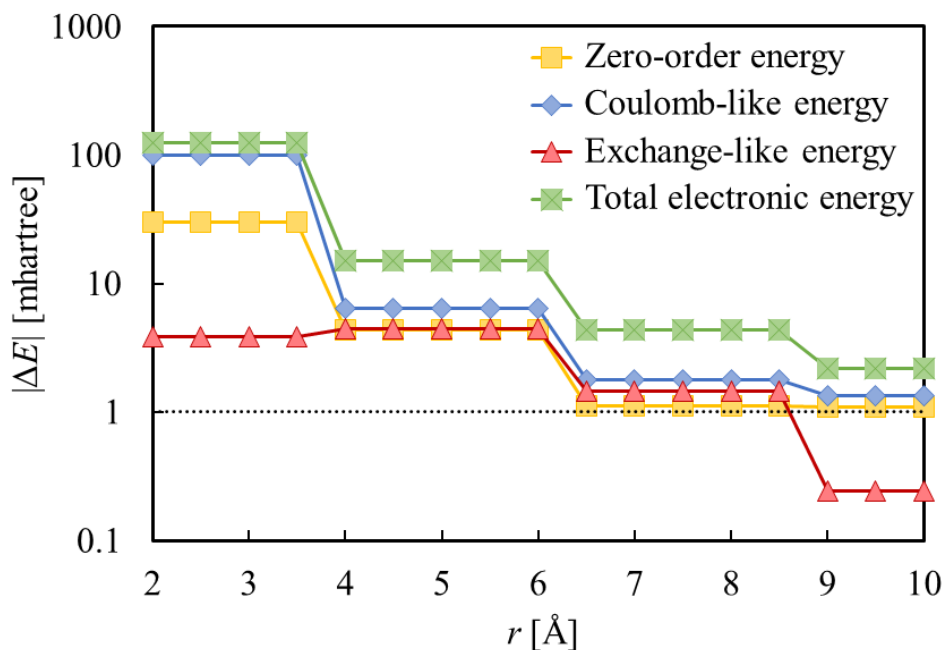


**Figure S1.** Time-course changes of  $r(\text{O}_1\text{-H}_1)$  and  $r(\text{O}_2\text{-H}_2)$  in BP(OH)<sub>2</sub> in gas phase.

## Buffer size dependence of ground-state energy

Figure S2 shows the buffer-size dependence of the ground-state energy of 10-acene.

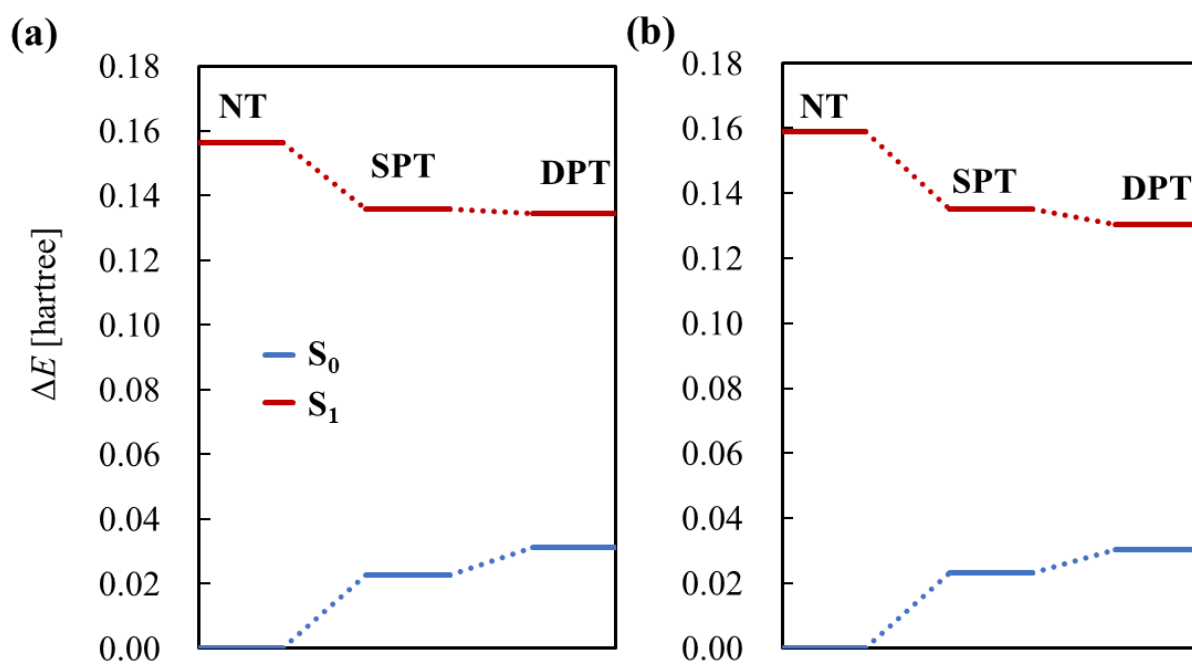
One benzene ring was used as a subsystem. Although required buffer size becomes larger, energy error between with DC and conventional methods is decreased by increasing the buffer size.



**Figure S2.** Dependence of the errors in the total electronic, zero-order Hamiltonian, Coulomb-like, and exchange-like energies, calculated using DC-LCDFTB from the conventional LCDFTB calculations on the buffer size ( $r$ ) of 10-acene.

## Potential energy of BP(OH)<sub>2</sub> in gas phase

The potential energies of NT, SPT, and DPT structures of BP(OH)<sub>2</sub> in gas phase were calculated by LCBLYP/aug-cc-pVDZ and LCDFTB/OB2. Figure S3 shows the energy differences  $\Delta E$  from the potential energy of NT structure in the  $S_0$  state. The  $S_0$ - $S_1$  energy difference and the energy difference between structures calculated by LCDFTB method provided the good agreement with LCBLYP.



**Figure S3.** Energy differences  $\Delta E$  from the potential energy of NT structure in the  $S_0$  state calculated by (a) LCBLYP/aug-cc-pVDZ and (b) LCDFTB/OB2.