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Theoretical Study on the Hole-Transport Property of Fullerene Hydrides $C_{60}H_2$ and $C_{60}H_4$

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Abstract Hole-transport property of $C_{60}H_2$ [1] and $C_{60}H_4$ [2] is discussed from the viewpoint of reorganization energy λ and hole-transfer rate constant k_{ht}, comparing with that of C_{60}. All synthesized isomers [3] of $C_{60}H_2$ and $C_{60}H_4$ have better hole-transport property than C_{60}. It is also revealed that the hole-transport property is closely related to the delocalization of HOMO.

Introduction Organic materials, which have lightness, flexibility, and are environmentally-friendly, are expected as essential parts of organic light-emitting devices (OLEDs) and organic field-effect transistors (OFETs). In these organic devices, hole mobility in the organic hole-transport material is one of the most important properties in the performance, therefore, the development of new material with good hole-transport property are matters of great urgency.

In the present study, we focus on the hole-transport property of fullerene hydrides $C_{60}H_2$ [1] and $C_{60}H_4$ [2], comparing with that of C_{60}. Hydrogenation has much influence on the hole-transport property of the C_{60} material because it removes electronic degeneracy of C_{60}^+. Potential utility of $C_{60}H_2$ and $C_{60}H_4$ as hole-transport material is discussed from the viewpoint of reorganization energy λ and hole-transfer rate constant k_{ht}.

Computational Method In Marcus theory, k_{ht} of a hole-transfer reaction between two equivalent molecules M(A) and M(B) is represented as

$$k_{ht} = \frac{4\pi^2}{h}\frac{H_{AB}{}^2}{\sqrt{4\pi\lambda k_B T}} e^{-\lambda/4k_B T}.$$

Thus, k_{ht} is mainly dependent on λ and the electronic coupling H_{AB}. Assuming that H_{AB} is the same for all molecules, smaller λ simply results in the larger k_{ht}. The calculation of λ and k_{ht} is performed by the density functional theory (B3LYP/6-311G(*d*)) using Gaussian 03.

Results and Discussions For selected 11 isomers of $C_{60}H_2$ and 9 isomers of $C_{60}H_4$ shown in Fig. 1, λ and k_{ht} were calculated on the assumption that H_{AB} is the same for all molecules

(See Fig. 2). $C_{60}H_4$ isomers overall tend to have good hole-transport property than C_{60}. Synthesized isomers [3] **0a** and **2a** of $C_{60}H_2$ have 20 % smaller λ than C_{60}, and k_{ht} of these are about 1.5 times as large as that of C_{60}. Also, isomer **1** of $C_{60}H_4$, which is the major product of the synthesis, has 50 % smaller λ, and its k_{ht} is about 3.3 times. These results indicate that some isomers of $C_{60}H_2$ and $C_{60}H_4$ have potential utility as hole-transport material.

Figure 3(a) shows HOMO of $C_{60}H_4$ isomer **1** with the largest k_{ht} and that of **2** with the smallest k_{ht}. It is found that **1** with more delocalized HOMO has larger k_{ht} than **2**. This result coincides with the intuitive picture of the hole hopping illustrated in Fig. 3(b).

Conclusions We studied the hole-transport property of $C_{60}H_2$ and $C_{60}H_4$ in terms of λ and k_{ht}. The major findings are as follows: i) the synthesized isomers of $C_{60}H_2$ and $C_{60}H_4$ have great potential as hole-transport material, ii) λ and k_{ht} are closely related to the delocalization of HOMO.

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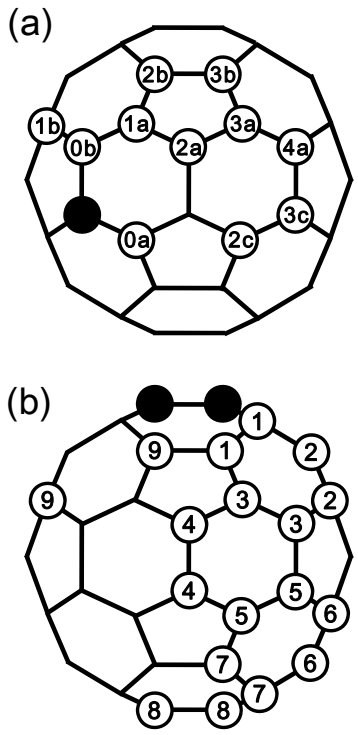

Figure 1. Isomers of (a) $C_{60}H_2$ and (b) $C_{60}H_4$. (a) Initial one H atom is placed at the filled circle, and the other H atom is at **0a**-**4a**. (b) Initial two H atoms are placed at the two filled circle, and the other two H atoms are at **1**-**9**.

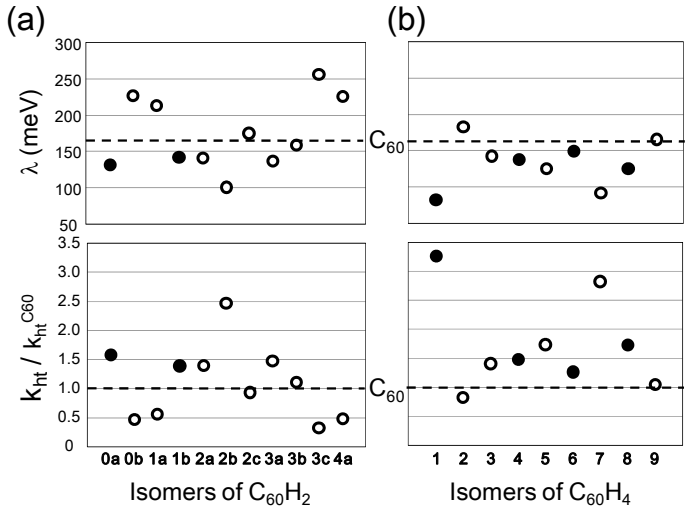

Figure 2. Calculated λ and k_{ht} / k_{ht}^{C60} (at 300 K) of (a) $C_{60}H_2$ and (b) $C_{60}H_4$ isomers. The filled circles show the values of the synthesized ones. The dashed lines show those of the original C_{60}.

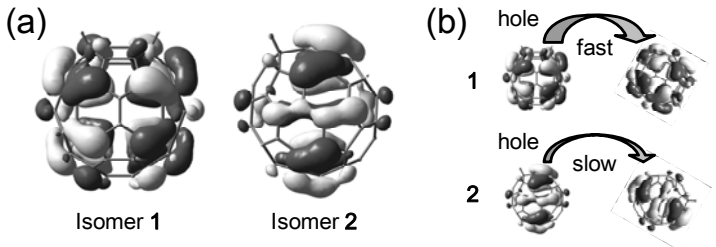

Figure 3. (a) HOMOs of $C_{60}H_4$ isomers, **1** and **2**. (b) Schematic illustrations of the hole-transport rate from the viewpoint of the delocalization of HOMO.

References **[1]** **K. Tokunaga**, S. Ohmori, H. Kawabata, and K. Matsushige, *Jpn. J. Appl. Phys.*, in press. **[2]** **K. Tokunag**, H. Kawabata, and K. Matsushige, submitted to *Jpn. J. Appl. Phys*. **[3]** K. Shigematsu and K. Abe, *Jpn. Patent*, P3161789, 2001-02-23.