Transactions of the Materials Research Society of Japan 25 [4] 1045-1047 (2000)

XPS Study of Si_xC_{60} Thin Films

Hideki Tanaka*, Jun Onoe, Toshiki Hara and Kazuo Takeuchi

RIKEN (The Institute of Physical and Chemical Research), 2-1 Hirosawa, Wako, Saitama 351-0198, Japan

*Fax: 81-48-462-4702, e-mail: htanaka@postman.riken.go.jp

The interaction between C_{60} and Si atoms was investigated using X-ray photoelectron spectroscopy (XPS) for the photo-irradiated Si-doped C_{60} thin film, $Si_x(C_{60})$, in order to establish a production method of Si-coated C_{60}, $C_{60}Si_{60}$. It was found that the concentration of Si atoms in the Si_xC_{60} thin film plays an important role in the formation process of $C_{60}Si_{60}$.

Keywords: Si-coated C_{60}; Si-doped C_{60} thin film; XPS; photo-induced reaction

1. INTRODUCTION

Si clusters have attracted much attention because of their potentials for new optical and electrical materials [1-5]. However, it is difficult to treat the Si clusters in actual processes because they are so reactive that their properties are affected by their surrounding atoms, molecules and surfaces.

Recently, Osawa and his co-workers theoretically predicted that $C_{60}Si_{60}$ (see Fig. 1) is more stable than the isolated C_{60} and Si_{60}, and that its energy levels of the frontier orbitals are similar to those of Si_{60} [6-8]. This suggests that it can be treated as a kind of single-shaped Si cluster. This molecule is, therefore, expected to be used as a functional nano-device when a method of its synthesis is established. In a previous work, we have investigated the interaction between C_{60} and Si atoms in a Si-layered C_{60} film from a starting point of the $C_{60}Si_{60}$ synthesis using X-ray photoelectron spectroscopy (XPS) [9]. While photo-irradiation process is found to play an important role for the $C_{60}Si_{60}$ formation, a variety of the C_{60}–Si interaction was observed in the same sample. In the present study, the authors formed Si-doped C_{60} thin film uniformly and examined the interaction between C_{60} and Si atoms using XPS.

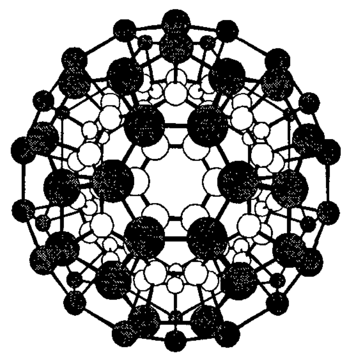

Fig. 1 Geometric structure of $C_{60}Si_{60}$. The open and closed circles represent C and Si atoms, respectively.

2. EXPERIMENT

The details of the experimental setup used in the present study have been described elsewhere [9-11]. The outline of experimental conditions is described here.

A CsI substrate of 20 mm diameter was heated to 100 °C for more than 1 hour in an ultrahigh vacuum chamber (less than 1×10^{-9} Torr) in order to evaporate water molecules adsorbed on the substrate. A Si-doped C_{60} thin film on the substrate was then prepared by the co-deposition of C_{60} molecules and Si atoms evaporated from a quartz crucible and from a Si rod sputtered by high-energy electrons, respectively. Afterward, the thin film was irradiated with UV-visible light of 1 $W\cdot cm^{-2}$ for 10 hours. After the substrate was taken out of the vacuum chamber, it was introduced into a box filled with helium gas and was moved to the XPS apparatus (VG ESCALAB Mk-II) within 1 hour. The substrate was introduced into a load-lock prechamber in the apparatus in order to be degassed under low pressure (10^{-3} Torr) for 30 minutes. The substrate was then transferred to the XPS-measurement chamber (5×10^{-10} Torr) and the XPS (MgKα) spectra of the sample were obtained.

3. RESULTS

Figure 1 (a) shows the C1s spectrum of the photo-irradiated Si-doped C_{60} film prepared by using 10 nA-Si-atom beam (sample A) in the binding-energy range of 280 – 290 eV. The binding energy of the C1s peak was estimated using the O1s (533 eV) peak as a reference. The dominant peak of the C1s spectrum is positioned at 284.2 eV. On the other hand, Fig. 1 (b) shows the C1s spectrum of the photo-irradiated Si-doped C_{60} film prepared by using 50 nA-Si-atom beam (sample B). In this spectrum, the dominant peak of the C1s spectrum is positioned at 284.8 eV.

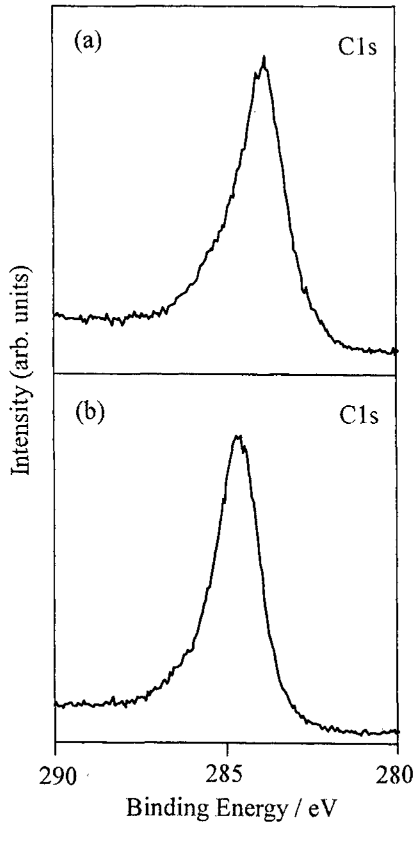

Fig. 2 The C1s spectra of the photo-irradiated Si-doped C_{60} thin films. The composition of the thin film is assigned to be about (a) $Si_2(C_{60})$ and (b) $Si_7(C_{60})$, respectively.

4. DISCUSSION

4.1 Composition of the Si-doped C_{60} thin film

The XPS intensity, I_A, of a certain core level for element A (A = C, Si) is expressed as [12]

$$I_A = \sigma_A \cdot I_X \cdot \lambda_A(E) \cdot N_A \cdot k_A(E) \ , \qquad (1)$$

where I_x is an intensity of the incident X ray, σ_A and N_A are an ionization cross section of the respective electronic level and a concentration of the element A, respectively, $\lambda(E)$ is the averaged escape depth of photoelectrons with a kinetic energy of E, and $k_A(E)$ is the spectrometer-transmission coefficient. When the two elements, C and Si atoms, are uniformly dispersed in the thin film, the same value I_X is considered to be applied to both elements. In this case, the concentration ratio, N_{Si} / N_C, can be obtained by

$$N_{Si} / N_C = (I_C / I_{Si})\cdot(\sigma_C / \sigma_{Si})\cdot(\lambda_C / \lambda_{Si})\cdot(k_C / k_{Si}). \qquad (2)$$

Using eq. (2), we found that the composition of the thin films is about $Si_2(C_{60})$ for sample A and $Si_7(C_{60})$ for sample B, respectively.

4.2 Interaction between C_{60} and Si atoms

Since the C1s binding energy is 285.0 eV for the C-C bonding in C_{60} molecule and 283.5 eV for the C-Si bonding in silicon carbide, the binding energy of the C1s photoelectron obtained for the Si-doped C_{60} thin films varies between these two values by depending on the magnitude of the interaction between C and Si atoms [13]. In fact, the chemical shift in the C1s peak from 285.0 eV for samples A and B is –0.8 eV and –0.2 eV, respectively. K. Sakamoto et al. has reported that chemical shift of –0.5 eV in the C1s spectrum indicates the onset of C_{60} molecules chemisorbed on Si surfaces [14]. This suggests that the large chemical shift observed for sample A arises from the chemical bond formation between the C_{60} and Si atoms (see Fig. 3(a)). On the other hand, the small chemical shift for sample B indicates Si atoms physisorbed to the C_{60} molecules. This implies that Si aggregates produced by diffusion process in highly concentrated Si-doped thin film are less chemically reactive for the C_{60} molecules than Si atoms (see Fig. 3(b)).

In summary, it is found that the concentration of Si atoms in the Si-doped C_{60} thin film becomes significant in the formation process of $C_{60}Si_{60}$.

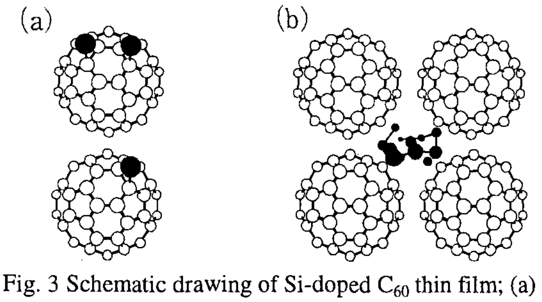

Fig. 3 Schematic drawing of Si-doped C_{60} thin film; (a) Si atoms (closed circle) chemically bound to C_{60} cage, (b) Si aggregates in C_{60} thin film.

5. ACKNOWLEDGEMENTS

The authors would like to thank A. Nakao of RIKEN for the XPS measurements. The present work was supported in part by a Grant-in-Aid for scientific research by the Science and Technology Agency of Japan and the Ministry of Education, Science and Culture of Japan.

REFERENCES

[1] E. C. Honea, A. Ogura, D. R. Peale, C. Félix, C. A. Murray, K. Raghavachari, W. O. Sprenger, M. F. Jarrold and W. L. Brown, *J. Chem.Phys.*, **110**, 12161-72 (1999).

[2] B. Dragnea and B. Bouguignon, *Phys. Rev. Lett.*, **82**, 3085-8 (1999).

[3] M. V. Wolkin, J. Jorne, P. M. Fauchet, G. Allan and C. Delerue, *Phys. Rev. Lett.*, **82**, 197-200 (1999).

[4] P. Mélinon, P. Kéghélian, B. Prével, V. Dupuis, A. Perez, B. Champagnon, Y. Guyot, M. Pellarin, J. Lermé, M. Broyer, J. L. Rousset and P. Delichère, *J. Chem. Phys.*, **108**, 4607-13 (1998).

[5] K. –M. Ho, A. A. Shvartsburg, B. Pan, Z. –Y. Lu, C. –Z. Wang, J. G. Wacker, J. L. Fye and M. F. Jarrold, *Nature*, **392**, 582-5 (1998).

[6] S. Osawa, M. Harada, E. Osawa, B. Kiran and E. D. Jemmis, *Fullerene Sci. Technol,.* **3**, 225-39 (1995).

[7] E. D. Jemmis, J. Leszczynski and E. Osawa, *Fullerene Sci. Technol,.* **6**, 271-81 (1998).

[8] H. Tanaka, S. Osawa, J. Onoe and K. Takeuchi, *J. Phys. Chem. B*, **103**, 5939-42 (1999).

[9] H. Tanaka, J. Onoe, T. Hara, A. Nakao and K. Takeuchi, *Mol. Cryst. Liq. Cryst.*, in press.

[10] J. Onoe and K. Takeuchi, *Phys. Rev. B*, **54**, 6167 (1996).

[11] J. Onoe, K. Takeuchi, K. Ohno, and Y. Kawazoe, *J. Vac. Sci. Technol. A*, **16**, 385 (1998).

[12] G. Ertl, J. Küppers, "Low Energy Electrons and Surface Chemistry", VCH (1985) pp. 65-85.

[13] A. Nakao, M. Iwaki, H. Sakairi, and K. Terasima, *Nucl. Instr. and Method B*, **65**, 352 (1992).

[14] K. Sakamoto, D. Kondo, Y. Ushimi, M. Harada, A. Kimura and S. Suto, *Phys. Rev. B*, **60**, 2579-91 (1999).

(Received December 17, 1999; Accepted March 11, 2000)