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In situ XPS study of Si-deposited C_{60} thin films

Hideki Tanaka*, Jun Onoe, 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 in a Si-deposited C_{60} thin film has been investigated by using *in situ* X-ray photoelectron spectroscopy (XPS). It is found that the charge-transfer complex $C_{60}Si_n$ was formed at room temperature after Si-atom beam deposition. The value of n for the charge-transfer complex $C_{60}Si_n$ was approximately up to 6.

Keywords: Si_{60}-coated C_{60}; Si-deposited C_{60} thin film; *in situ* XPS; charge-transfer complex

1. INTRODUCTION

Si clusters have attracted much attention as new optical and electrical materials [1-5]. However, it is difficult to handle the Si clusters in actual processes, because they are so unstable that their properties are easily affected by their surroundings.

Figure 1 shows the geometric structure of Si_{60}-coated C_{60} [6-8]. This has been theoretically predicted to be stable as same as C_{60} and to behave like a single-shaped Si_{60} cluster, which is expected to be used as a functional nano-material when its synthesis method is established. We have previously investigated the reaction of C_{60} with Si atoms using *ex situ* X-ray photoelectron spectroscopy (XPS) [9, 10]. Although it was found that the reaction proceeds in the Si-deposited C_{60} thin film, it was hard to analyze the chemical states of the Si atoms in the presence of Si oxides. In the present study, we performed *in situ* XPS measurements for the Si-deposited C_{60} thin film and discussed the stoichiometry of $C_{60}Si_n$ compounds quantitatively. Observed chemical shift of C 1s and Si 2p spectrum was compared with that of those spectra for C_{60} adsorbed Si surface [11].

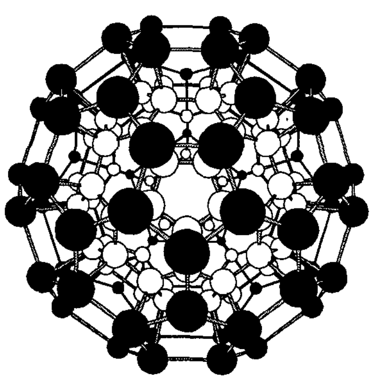

Fig. 1 Geometric structure of Si_{60}-coated C_{60}. The open and closed circles represent C and Si atoms, respectively.

2. EXPERIMENT

The details of the experimental setup for XPS measurement have been described elsewhere [12]. In this study, we installed a sample-preparation chamber to perform *in situ* XPS measurement. The outline of experimental conditions is described here.

The sample-preparation chamber was equipped with a turbo molecular pump, a C_{60} vapor source, and a Si atom-beam source (Omicron, EFM3), and was isolated from the XPS analysis chamber via a gate valve. The base pressure of the sample-preparation chamber and the XPS analysis chamber was kept at less than 1 × 10^{-7} Pa.

A stainless steel substrate with 20 mm in diameter was introduced into the sample-preparation chamber. A pristine C_{60} thin film was formed on the substrate by sublimation of C_{60} powder. Thereafter, the film was partially exposed to Si-atom beam with about 10 mm in diameter in order to form a pristine C_{60} thin film and a Si-deposited C_{60} thin film on the same substrate. The current of the Si-atom beam was adjusted to 20 nA by conventional flux monitor. The sample thus formed was transferred into the XPS analysis chamber, and the XPS (MgKα) spectra of the film were obtained *in situ*. The escape depth of the C 1s photoelectrons for this apparatus was estimated to be about 1.8 nm [13]. The binding energy of the spectrum was determined using the C 1s (284.7 eV) peak of the pristine C_{60} thin film as a reference [11]. Analysis of the spectrum was carried out by Fisions analysis software package (ECLIPS).

The deposition rate of the Si_x-deposited C_{60} thin film was estimated by

$$x = N_{Si} / (N_C / 60) \tag{1}$$

where N_{Si} and N_C are the number of Si and C atoms, respectively. From the observed spectra, we found that the x is 21 ± 4 for 1 hour deposition of Si atoms on one C_{60}.

3. RESULTS AND DISCUSSION

3.1 Chemical shift of Si 2p spectrum

The Si 2p spectra of the Si-deposited C_{60} thin film are shown in Fig. 2. Total exposure time of the Si-atom beam is represented on the right side of each spectrum. The dotted line represents the peak position for a clean Si surface (99.6 eV). The binding energy of the peak position for the spectra decreases as the exposure time increases.

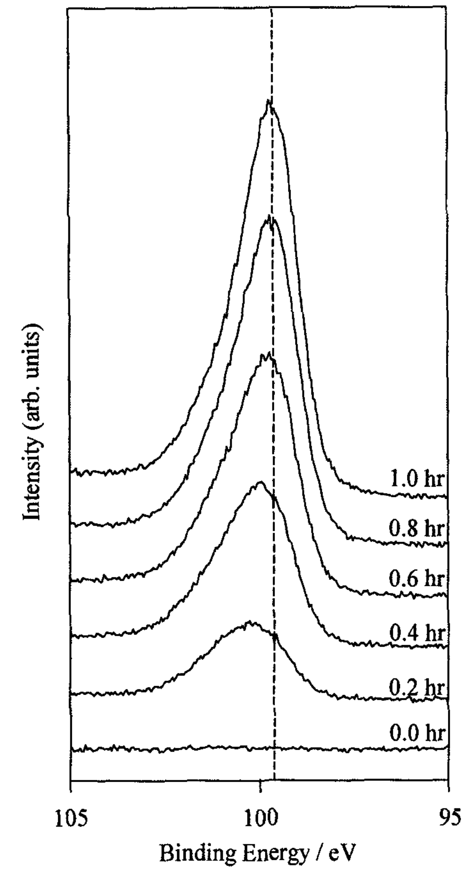

Fig. 2 The Si 2p spectra of Si-deposited C_{60} thin film. Total exposure time of the Si-atom beam is represented on the right side of the spectrum, respectively. The dotted line represents a peak position for a clean Si surface.

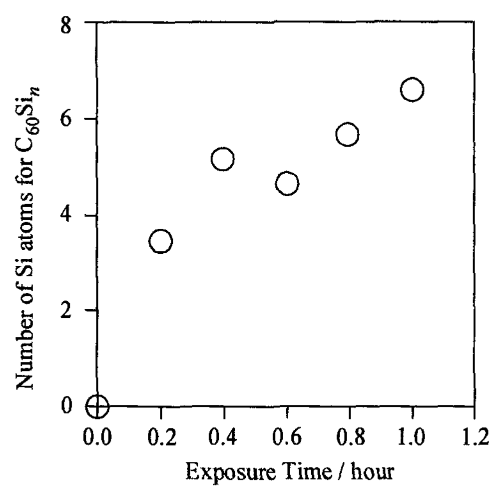

Fig. 3 Number n of Si atoms for the charge-transfer complex $C_{60}Si_n$ is plotted as a function of exposure time for the Si-atom beam.

In the present XPS system, the FWHM value of 2.0 ± 0.2 eV was suitable for peak fitting of Si 2p spectrum [14]. Using this value, all the spectra can be analyzed by considering two peaks at 100.2 eV and 99.6 eV. An increase of the peak at 99.6 eV indicates the formation of Si aggregates. The chemical shift of +0.6 eV (= 100.2 − 99.6) is comparable to that for C_{60} adsorbed on the Si surface at 670 K (+0.4 ~ +0.5 eV), which has been reported by Suto et al [11]. According to their report, they concluded that the charge transfer from the Si surface to C_{60} molecules takes place. In a similar manner, the chemical shift of −0.4 eV was observed in the C 1s spectrum. This shift is also almost equal to that for the charge-transfer complex $C_{60}Si_n$ formed on the C_{60}-adsorbed Si surface at 670 K [11].

3.2 Charge-transfer complex $C_{60}Si_n$

The Si-deposited C_{60} thin film is composed of the charge-transfer complex $C_{60}Si_n$ and the Si aggregates. The value of n was estimated as follows. The number of Si and C atoms for the complex was estimated from the intensity of the Si 2p and C 1s peaks shifted by charge-transfer in their spectra, respectively. Then we used these numbers thus estimated in eq.(1), instead of the N_{Si} and N_C. Figure 3 shows the number n of Si atoms for the charge-transfer complex $C_{60}Si_n$ as a function of exposure time of the Si-atom beam. The number n increases rapidly with the exposure time, and tends to reach the asymptotic limit of approximately 6.

4. SUMMARY

It is found that the charge-transfer complex $C_{60}Si_n$ was formed for Si-deposited C_{60} film at room temperature, and the value of n for the charge-transfer complex $C_{60}Si_n$ with sufficient Si atoms is approximately 6.

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