DOI: 10.1017/S1431927606064725
Microsc Microanal 12(Supp 2), 2006
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Radiation Damage of C_{60} Single Molecules Encapsulated in Carbon Nanotube

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Radiation damage study of fullerene (C_{60}) single molecules encapsulated in single-wall carbon nanotubes (SWCNT) has been attempted by high-resolution transmission electron microscopy (HR-TEM) at 120 kV. In order to optimize the condition of HR-TEM imaging, coalescence process of C_{60} peapod is quantitatively analyzed with accumulated electron dosage. Effects on coalescence at different temperature (93K, 298K, 550K; FIG.1), filling space (intermolecular distance; FIG.2), and differences from crystalline transformation (FIG. 3) are carefully analyzed. At 100 kV, required electrons for 1 incident of ionization and atomic displacement are estimated to 2.4×10^{-3} and 27.2 C/cm^2, respectively [1]. In the present study, the minimum electron dose required for one image is estimated to be 0.2 to 1.0 C/cm^2 so that the imparted electron is still large enough to cause ionization.

In FIG.1, C_{60} molecules in peapod preserve its structure at low temperature (a)-(d). Assuming ionization and knock-on probabilities are temperature-independent, low rate of coalescence would be due to low mobility of C_{60} (cage effect) and low thermal vibrations. At room temperature (e)-(h) or high temperature (i)-(l), molecules are more reactive as a result of high mobility of C_{60}. When C_{60} molecules invade their equilibrium intermolecular distance, high rate of coalescence seems to occur. Unlike nanotube [2], recombinations of displaced atoms are very little even at 550K.

High mobility of an isolated C_{60} may cause fast coalescence with low electron irradiation. Isolated C_{60} molecules (FIG.2(a)-(d)) dimerize with smaller electron dosage, while closely-packed C_{60} molecules (FIG.1(e)-(h)) preserve their structure to much higher electron dosage. A relaxation of ionization may be increased in a closely-packed C_{60} array, which stabilizes the C_{60} structure under the electron radiation. Ionization may be localized in segmented C_{60} molecules and simultaneous ionization on adjacent C_{60} molecules may cause coalescence (dimerization).

In the bulk C_{60} crystal (FIG.3), damage rate of each molecule is almost same as that of C_{60} peapod. Nevertheless the lattice planes of C_{60} crystal are observable at higher electron radiation, which does not mean no damage in C_{60} molecules. Most of C_{60} molecules lose original structure but barely preserve original lattice distance. Lattice plane perpendicular to the crystal edge remains at high electron irradiation. The result suggests that coalescences of C_{60} molecule transmit from damaged area to inner part of the crystal along a certain direction.

In analogous to the study of C_{60} peapod and crystal, it is expected that HR-TEM imaging of single molecule in SWCNT becomes attainable by applying low temperature, and preparing closely packed molecules or a completely isolated molecule. At low temperature, mechanical stability becomes critical so it would be a compromise of radiation damage and high-resolution stability.

References

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[2] K. Urita, *et al. Nano Lett.* (2004) 2451-2454.

https://doi.org/10.1017/S1431927606064725 Published online by Cambridge University Press

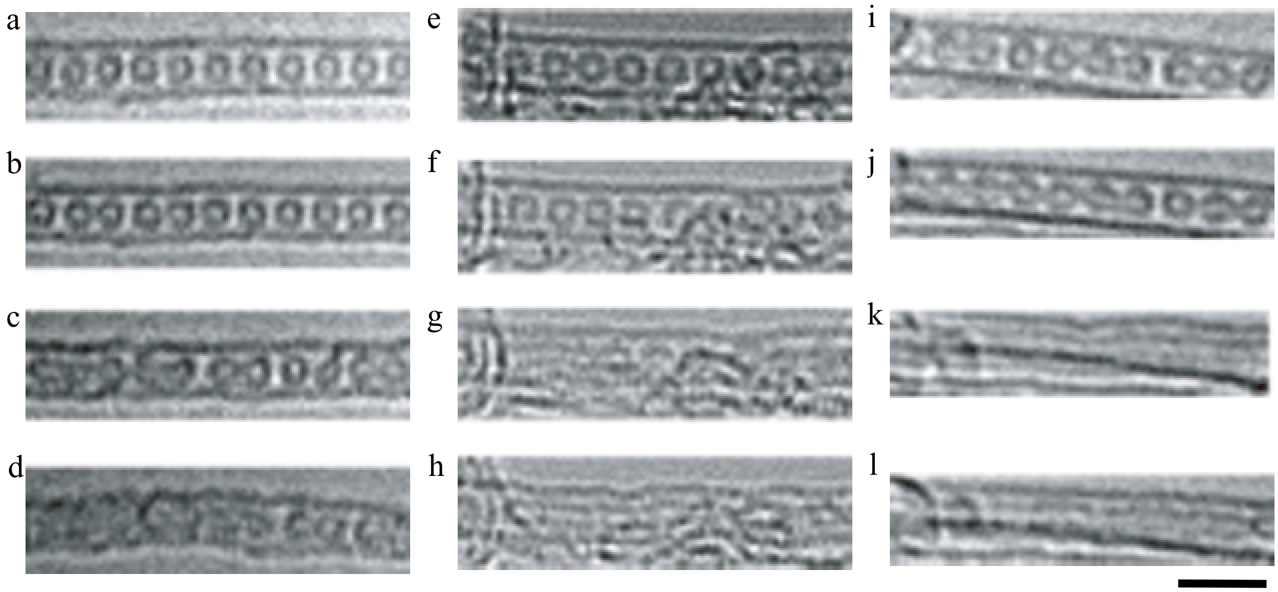

FIG. 1. Coalescence of C_{60} molecules in peapod at different temperature: 93K (a)-(d), 298K (e)-(h), and 550K (i)-(l). Electron dosage imparted to the sample is 5, 70, 481, and 881 Ccm^{-2} from top to bottom, respectively. Scale bar is 2nm.

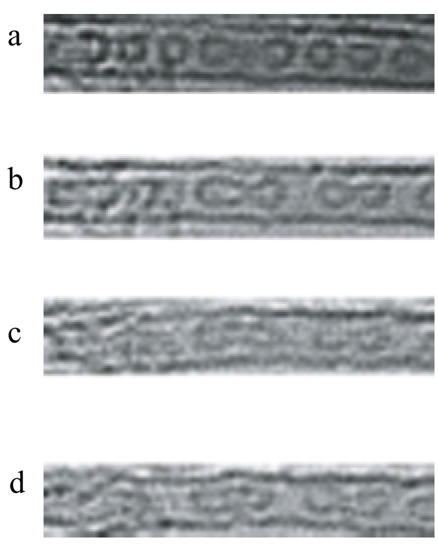

FIG. 2. High sensitivity to the radiation dose is observed in C_{60} peapods with a large filling space (intermolecular distance) at room temperature. Electron dosage imparted to the sample is same as Fig.1. Scale bar is 2nm.

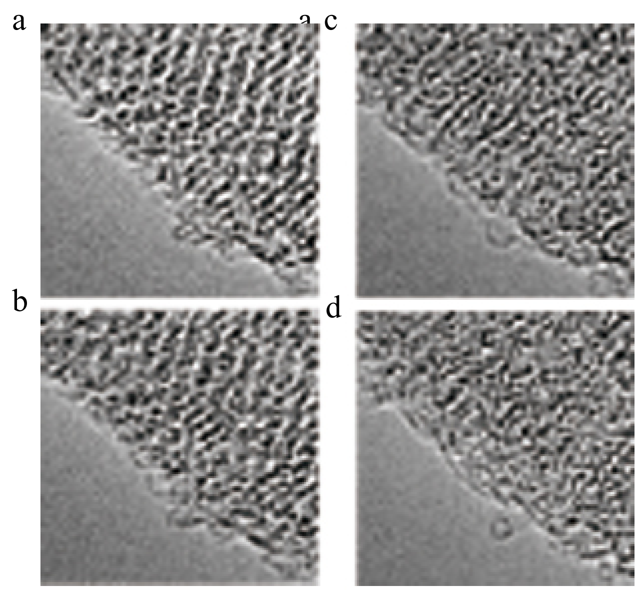

FIG. 3. Radiation damage of C_{60} crystals at room temperature. Electron dosage imparted to the sample is same as Fig.2. Scale bar is 2nm.

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