Chirality distributions of grown single-walled carbon nanotubes assigned by photoluminescence spectroscopy

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1. Introduction

In order to realize single-walled carbon nanotube (SWNT) chiral selective growth, it is important to elucidate the mechanism of SWNT chirality (n,m) selectivity. For this purpose, an accurate evaluation method for chirality distribution of grown SWNTs without post-growth processing and liquid-dispersion of SWNTs is indispensable. Here, we used photoluminescence (PL) spectroscopy to directly measure the chirality distributions of individual SWNTs suspended on a pillar-patterned substrate.

2. Experimental

The pillar-patterned substrate had 1296 pillar pairs with a height of 10 μm and spacing of 5 μm. SWNTs was synthesized directly on the pillar substrate using ethanol chemical vapor deposition method [Co catalyst with a thickness of 7.8 pm, 750-870C° in temperature, 2-14 min in growth time]. The excitation wavelength λ_{22} for PL spectroscopy was in the range of 730–850 nm, and the detection wavelength λ_{11} was set to approximately 1000–1500 nm. The chirality of SWNT can be determined from the combination of λ_{22} and λ_{11}. In our emission/detection wavelength ranges, 22 types of SWNTs can be detected.

3. Results and Discussion

We evaluated the tendency of SWNT chirality assigned by PL map. The pillars searched were 7,776 (12 substrates). The number of chirality-assigned SWNTs was as large as 248 in total and 16 chirality types with the chiral angle ranging from 0° to 28.05° were detected. The percentages of SWNTs that could be assigned were 1.6%. We fitted the diameter distribution data with Gaussian functions as shown in Fig. 1a. The diameter distribution extends over the range of standard deviation σ = 0.09 nm centered at the mean value μ_D = 1.11 nm. On the other hand, Fig. 1b shows the chiral angle distribution. A straight-line fitting was performed assuming a screw dislocation model.[1] SWNTs with smaller chiral angles have lower yield. Therefore, the growth of SWNTs mostly depends on the chiral angle. We have confirmed that near armchair SWNTs are more likely to grow as suggested by the screw dislocation model.[1]

Figures 1c show chirality distributions of grown SWNTs on the (n, m) diagram. From these figures, the diameters and chiral angles of SWNTs likely to grow are seen at a glance. They distribute around (9,7), (9,8) and (10,5) at μ_D = 1.11 nm and $\theta \approx 30°$. Armchair SWNTs could not be detected by PL because they are metallic. The only zigzag SWNT that could be detected was (14,0). However, the probability was extremely low.

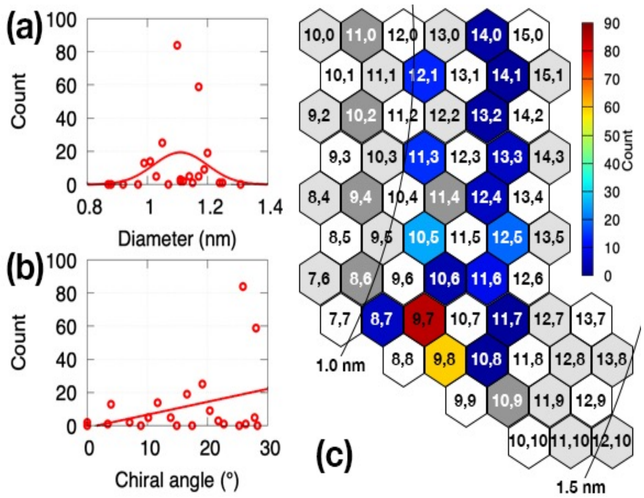

Fig. 1 (a) The diameter and (b) chiral angle distributions of SWNTs whose chirality could be assigned by PL measurement. Red line in (a) shows Gaussian fittings. (c) Counts and chirality distributions of grown SWNT. The six-membraned rings of dark gray, white, and gray indicate SWNT chiralities of unobserved, metallic, and out of our measurement range, respectively.

Furthermore, when higher yield chiralities are selected, the chiral angle distribution with a peak at near-armchair SWNTs is well fitted with the model taking the thermodynamic effect at the SWNT-catalyst interface into the kink growth-based kinetic model[2], i.e., a(30° - θ) exp[b(30° - θ)], where a and b are fitting parameters as seen in Fig. 2. The higher yield for (9,7) than (9,8) is consistent with the model.

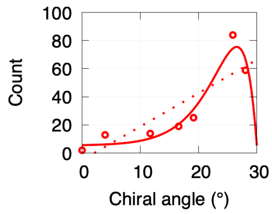

Fig. 2 The chiral angle distributions of higher yield SWNTs.

3. Conclusions

Our quantitative and statistical data provide new insight into SWNT growth mechanism as well as experimental confirmation of theoretical predictions.

Acknowledgements

This work was supported by JST, CREST Grant Number JPMJCR18I5, Japan.

References

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