Tracing individual growth process of single-walled carbon nanotubes by digitally coding isotope labels

Keigo Otsuka[1,2], Shun Yamamoto[1], Taiki Inoue[1], Rong Xiang[1], Shohei Chiashi[1], and Shigeo Maruyama[1,3]

[1] *The University of Tokyo, 7-3-1 Hongo, 113-8656, Japan*
[2] *RIKEN, 2-1 Hirosawa, 351-0198, Japan*
[3] *AIST, 1-2-1 Namiki, 305-8564, Japan*

Arrays of purely semiconducting single-walled carbon nanotubes (SWNTs) attract a significant attention as alternative materials of silicon for high-performance electronics. Despite recent progress in controlled synthesis of SWNTs [1], the growth process leaves some mysteries due to their size, variety and system complexity. Although thermodynamic and kinetic control is important for the chirality-controlled synthesis, it is controversial, for example, whether the growth rate of SWNTs depends on their chirality [1,2]. Usual *in situ* and *ex situ* measurements often missed a variation of grown SWNTs and the time evolution of the growth, respectively. Here we present a method to analyze the growth behavior, *e.g.* growth rate, incubation and lifetime, of individual SWNTs with various lengths by embedding digitally coded isotope labels.

SWNTs were grown from iron nanoparticles patterned in stripes on quartz substrates [2]. During the synthesis of SWNTs, binary-like codes of isotopic carbon were incorporated by periodically introducing the pulse of ^{13}C ethanol with three different ratios (Fig. 1a), and then detected by Raman mapping. A Raman intensity map along an SWNT (Fig. 1b) shows G-band downshifts in three different levels. We found that after various lengths of incubation, most SWNTs monotonically elongated until abrupt termination (Fig. 1c). This indicates the catalytic activity of nanoparticles did not change with time. Digital coding of isotope labels also offered opportunities to link modulation of growth conditions, such as temperature and carbon concentration (Fig. 1d), with changes in growth behavior along individual SWNTs. Similar analyses for the growth with a variety of catalysts and conditions would significantly improve the synthesis of SWNTs once combined with other characterization methods.

[1] S. Zhang *et al*., Nature **543**, 234 (2017).

[2] T. Inoue, D. Hasegawa, S. Chiashi, and S. Maruyama, J. Mater. Chem. A **3**, 15119 (2015).

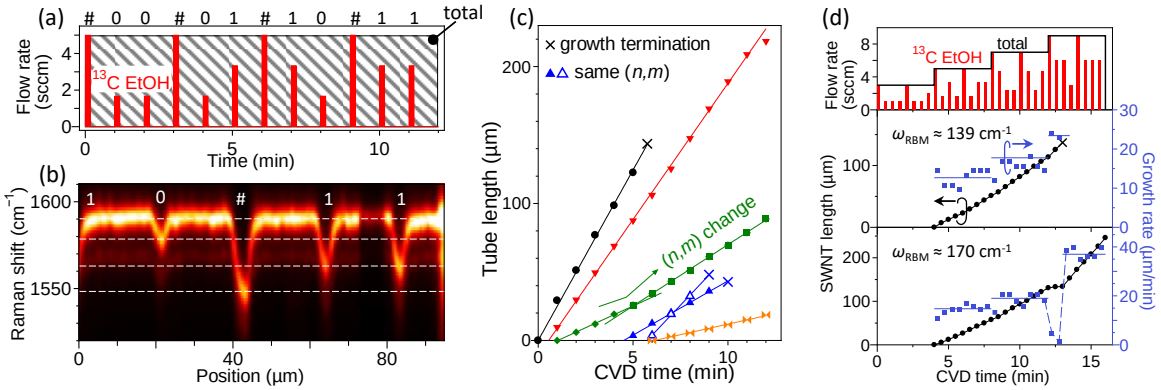

Fig.1 (a) Flow rate of ^{13}C ethanol (bars) for digital isotope coding with constant flow rate (5 sccm) of total ethanol. (b) G-band intensity map along an SWNT. (c,d) Time evolution of SWNT lengths with a constant growth condition (c), and with elevated ethanol flow rates (d).