カーボンナノチューブのリチウムイオン電池の電極材料応用

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１．　研究課題

Single-wall carbon nanotube (SWCNT), as an important type of carbon nanotubes (CNTs), consists of a single graphene seamlessly wrapped into a cylindrical tube. Owing to their lightweight and high electrical conductivities, CNTs have exhibited practical application potentials in batteries, capacitors, solar cell etc., In this research, we focus on yielding SWCNTs with high using a self-developed fluidized-bed chemical vaper deposition method.

２．　主な研究成果

(1) Deposition of catalyst precursor films by sputtering

In this research, ZrO_2 beads were chosen as substrates due to their highly stable chemical stability and sufficient surface area for the growth of CNTs. A home-made drum-sputtering facility is used to sputter uniform catalyst precursor films (Al and Fe in this research) on ZrO_2 beads, and an octagonal drum is newly designed to promote the radially and axially rotation of beads in the drum. We firstly measured the deposition rate of Al layer and Fe layer by performing sputtering at different power.

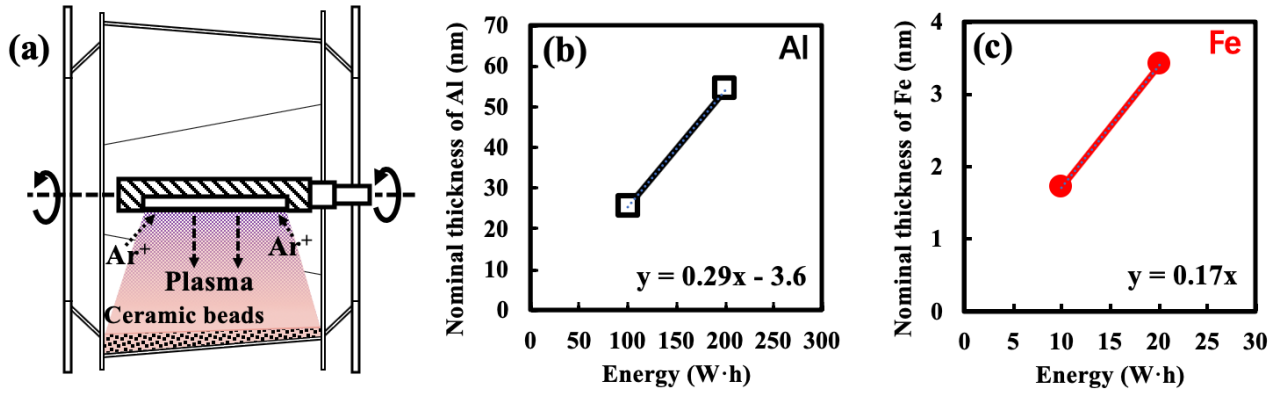

Fig. 1 Schematic diagram of drum-sputtering facility and the measured deposition rate. (a) Schematic image of designed octagonal drum. Deposition rate of (b) Al layer sputtered at 120 W and (c) Fe layer sputtered at 20W performed by adding 100 g beads under Ar atmosphere for 60 min.

X-ray fluorescence spectrometer results show that the average deposition rate was 0.78 nm/min for Al layer and 0.06 nm/min for Fe layer. Therefore, we confirmed the deposition time

of 15 nm Al bottom layer is 19 min and that of 1 nm Fe top layer is 17 min. We then carried out the CVD synthesis of SWCNT using ZrO_2 beads with target thickness of Al and Fe.

(3) Synthesis of SWCNTs by fluidized-bed CVD.

We optimized the synthesis condition in detail by increasing annealing time from 10 to 30 min. Fig.2 a-f showed the morphology and detailed structure of synthesized SWCNTs, and their average diameter is summarized in Fig. 2i. The higher SWCNT arrays grew from the surface of beads with the prolonged annealing time, and the length of 0.3 mm was achieved at the annealing time of 30 min. The typical TEM images and SWCNT content are summarized in Fig. 2d-f. Most of the observed CNTs are single-wall with an average diameter of 2.8-3.0 nm, while the content of SWCNTs reduced from 95 % to 83 % because the Fe catalyst particles coarsen into larger ones through surface diffusion. In Fig. 2h, we summarized the yield and specific surface area of SWCNTs. The yield of SWCNTs were evaluated by measuring the mass change before and after the combustion of SWCNTs on ZrO_2 beads. The highest yield of 14 mg_{CNT}/g_{beads} and specific surface area of 1221 m^2/g is achieved at the annealing time of 20 min, suggesting the optimum synthesis condition to form catalyst particles with higher number densities. Additionally, the Raman spectra showed that the typical D-band peak at 1350 cm-1 and G-band peak at 1580 cm^{-1} for CNTs were observed. The radical breathing mode (RBM) at 100-300 cm^{-1} were observed, indicating the existence of SWCNTs again. The intensity ratio of G-band peak and D-band peak (I_G/I_D) is ~5 for the samples prepared after 20 min annealing. These results show that high quality SWCNTs with large SSA can be produced using our fluidized-bed CVD method.

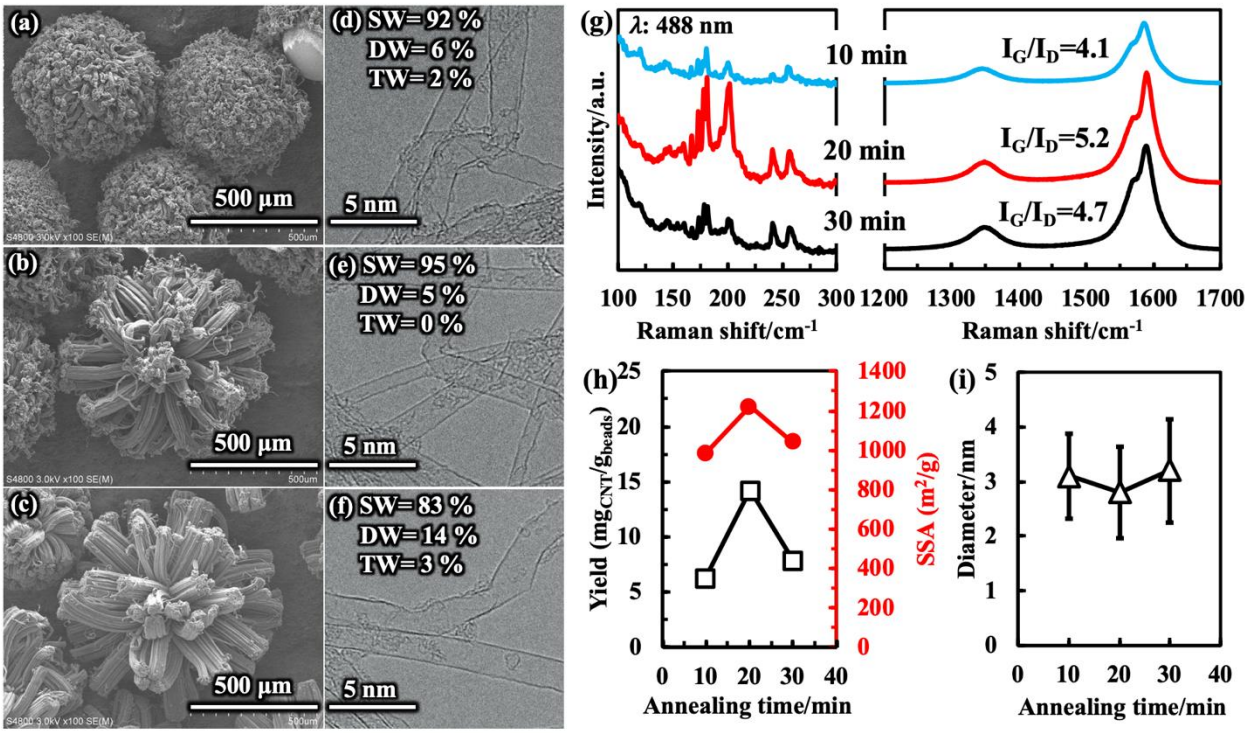

Fig. 2 SWCNT arrays synthesized by FBCVD. (a-c) scanning electron microscope images, (b-f) transmission electron microscope images, (g) Raman spectroscopy, (h) yield and specific

surface area, and (i) average diameter of SWCNTs synthesized at the annealing time of 10, 20 and 30 min.

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４． 研究業績

4.1 学術論文

[1] T. Liu, K.-C. Kim, B. Lee, S. Jin, M. Lee, **M. Li**, S. Noda, S. S. Jang*, and S. W. Lee*, Enhanced lithium storage of an organic cathode via bipolar mechanism, ACS Appl. Energy Mater., in press.

4.2 総説・著書　特になし

4.3 招待講演　特になし

4.4 受賞・表彰　特になし

4.5 学会および社会的活動

[1] **Mochen Li**, Risa Maeda, Toshio Osawa, Hisashi Sugime, and Suguru Noda, "Facile catalyst deposition using mist for fluidized-bed production of sub-millimeter-long carbon nanotubes," Guadalupe Workshop IX: Workshop on Nucleation and Growth Mechanisms of Single Wall Carbon Nanotubes, Fredericksburge, TX, USA, Apr. 17, 2019 (poster).

[2] **Mochen Li**, Risa Maeda, Toshio Osawa, Hisashi Sugime, and Suguru Noda, "Facile catalyst deposition using mist for fluidized-bed production of sub-millimeter-long carbon nanotubes," NT19: International Conference on the Science and Application of Nanotubes and Low-Dimensional Materials, (BoA p103) P067, Wurzburg, Germany, Jul. 22, 2019 (poster).

[3] 立川 明林，**Li Mochen**，杉目 恒志，大沢 利男，野田 優 「流動層による有機金属蒸気供給と触媒担持および長尺カーボンナノチューブの合成」, 化学工学会第 85 年会，PD372， 関西大学千里山キャンパス，大阪府吹田市，2020 年 3 月 17 日(poster).

[4] 白川 寛人，**Li Mochen**，杉目 恒志，大沢 利男，野田 優 「流動層による金属硝酸塩ミストからの触媒担持と長尺カーボンナノチューブの流動層合成」, 化学工学会第 85 年会，PC261， 関西大学千里山キャンパス，大阪府吹田市，2020 年 3 月 16 日(poster).

５． 研究活動の課題と展望

We will focus on the synthesis of SWCNTs (specific surface area>1000 m^2/g) and expect to increase the carbon yield to 15 at% using fluidized-bed CVD. Besides, quick deposition of metalorganic catalyst precursor film is expected to be finished in 3 min and FWCNTs (number of wall<3) are expected to be synthesized at the carbon yield larger than 30 at% using C_2H_2 as carbon source.