

# Three dimensional arrangements of carbon nanotubes by dry release approach

Zhaoqian Liu, Hui Li, Jing Yang, Zhijun Qiu, Ran Liu\*

State Key Lab of ASIC and System, Fudan University, Shanghai, 200433, China  
rliu@fudan.edu.cn

Gaoshan Huang, Yongfeng Mei\*

Department of Materials Science, Fudan University, Shanghai, 200433, China  
yfm@fudan.edu.cn

Carbon nanotubes (CNTs), owing to their outstanding electrical, optical, mechanical and thermal properties, have attracted numerous attentions and have been applied in wide realms. However, to our knowledge, to arrange carbon nanotubes in three-dimension has not been reported yet. One simple way to get some specific structures is to bend the flat membrane covered with CNTs by rolling approach.<sup>1</sup> In this work, the method of dry release<sup>2</sup> is employed for the reason that traditional rolled up technology requires multiple processes and undergoes some wet chemicals which might flush away CNTs.

Figure 1 schematically shows the fabrication steps: a) after spinning coating of PMMA on Si substrate, pre-strained layers of 32 nm SiO<sub>x</sub> and 8 nm SiO<sub>2</sub> were evaporated via electron beam evaporation; b) a drop of aqueous solution of CNTs was dipped on the surface and gradually dried at room temperature; c) heating of the sample at 400 °C for 2 h in N<sub>2</sub> atmosphere was carried out in a furnace. The heating process, herein, renders the removal of the sacrificial layer PMMA and, thus, stimulates the release and rolling of pre-strained layers into tubular structures, as reported in previous work.<sup>2</sup>

Figure 2 presents some images of morphology of CNTs and rolled up micro tubes. Well distributed CNTs on flat films after the heating process (in (a)) indicated that CNTs can firmly adhere to the deposited films and will not be blew all away after all the processes. Concerning the dispersant in the aqueous solution of CNTs, the tube wall will not be perfectly smooth and not be attached by any CNTs as can be seen in (b). However, if CNTs can be uniformly distributed in the dispersant and on the deposited SiO<sub>2</sub> surface, a novel tubular structure with erected CNTs can be fabricated as can be seen in (c).

Figure 3 displays several spectra of CNTs to see the influence the rolled up films on CNTs. The spectrum of as dipped sample shows typical spectral features of semiconducting single wall carbon nanotubes. After the heating process, the spectra of CNTs on unrolled films and rolled up tubes are recorded as can be seen in Figure 3. Though the peak positions of G band did not shift, the half width of the peaks first get wider after heating and, subsequently, narrowed down in the rolled up tubes. The inset in Figure 3 shows the spectra of dipped

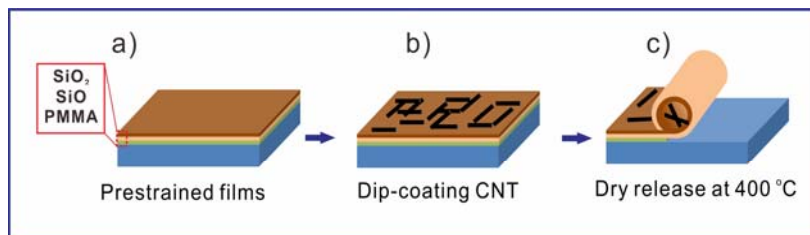
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<sup>1</sup> Y. F. Mei, G. S. Huang, A. A. Solovev, E. B. Urenã, I. Mönch, F. Ding, T. Reindl, R. K. Y. Fu, Paul K. Chu, and O. G. Schmidt, *Adv. Mater.* 20 (2008) 4085–4090.

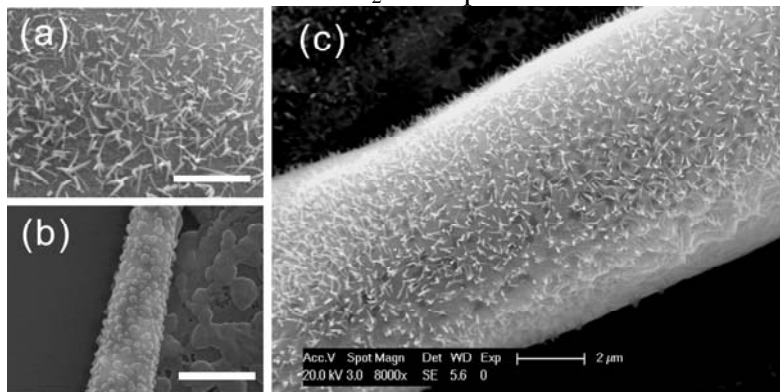
<sup>2</sup> J.X. Li, J. Zhang, W. Gao, G.S. Huang, Z.F. Di, R. Liu, Joseph Wang ,and Y.F. Mei, *Adv. Mater.* 25 (2013) 3715-3721.

CNTs before and after heating at 100 °C on a hotplate for 20, 60, 100 min. The spectra barely shift, indicating that the elimination of solvent at 400 °C merely affected the spectra. The details and mechanisms are still under investigations.

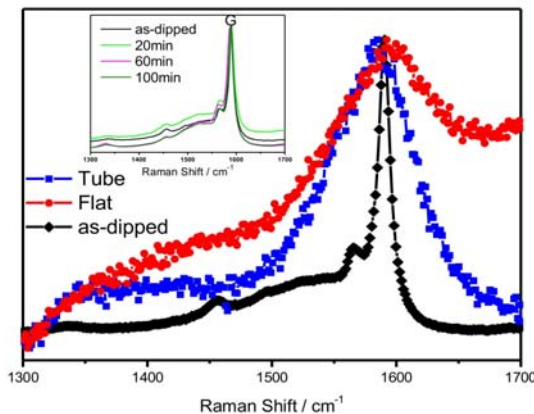
In summary, we have successfully rolled up CNTs by using dry release method. It would be much harder to roll up CNTs with traditional rolled up technique for the fact that wet chemicals and extra drying process may flush CNTs away. The effect of rolled up tubes on the Raman spectrum of CNTs is still under research, nonetheless, it can be anticipated that this novel tubular structure is of great potential in optics, electrics, photoelectrics, bio-technology.



*Figure 1: Schematics of rolling carbon nanotubes (CNTs):* After deposition of pre-strained films of  $\text{SiO}_x$  and  $\text{SiO}_2$  on a sacrificial layer PMMA (a), CNTs are dip-coated (b). CNTs, together with strained films, are rolled up (c) when heated at 400 °C for 2 h in  $\text{N}_2$  atmosphere in a furnace.



*Figure 2: SEM images of well distributed CNTs on flat films (a), a micro-tube rolled with dispersant (b), a micro-tube attached by CNTs (c). Scale bars are 2  $\mu\text{m}$ .*



*Figure 3: Different Raman spectra from CNTs on flat strained films and micro tubes after heating 2 h at 400 °C in  $\text{N}_2$  atmosphere. The inset shows the CNTs Raman spectra before and after heating at 100 °C for 20, 60, 100 min.*