Electronic Supplementary Material (ESI) for RSC Advances.

Supplementary information 1 (S1) : The microstructures of synthesized CNTs from liquid feeds

The CNT appeared in Fig. 2a is a MWCNT with amorphous carbons. We chose this one to demonstrate the sulfur distribution effect on the formation of a CNT even in a single catalyst particle. We have also observed many DWCNTs and TWCNT at the same sample, and present here two HRTEM photographs which clearly show the parallel graphitic walls with small amount of amorphous carbons. The number of walls can be counted easily from these Figures, and they are DWCNTs (Fig.S1-1) and TWCNTs (Fig. S1-2). It is very difficult to synthesize only one type of CNTs. Different types of CNTs are frequently synthesized with narrow or wide distribution depending upon the skill of an experimenter.

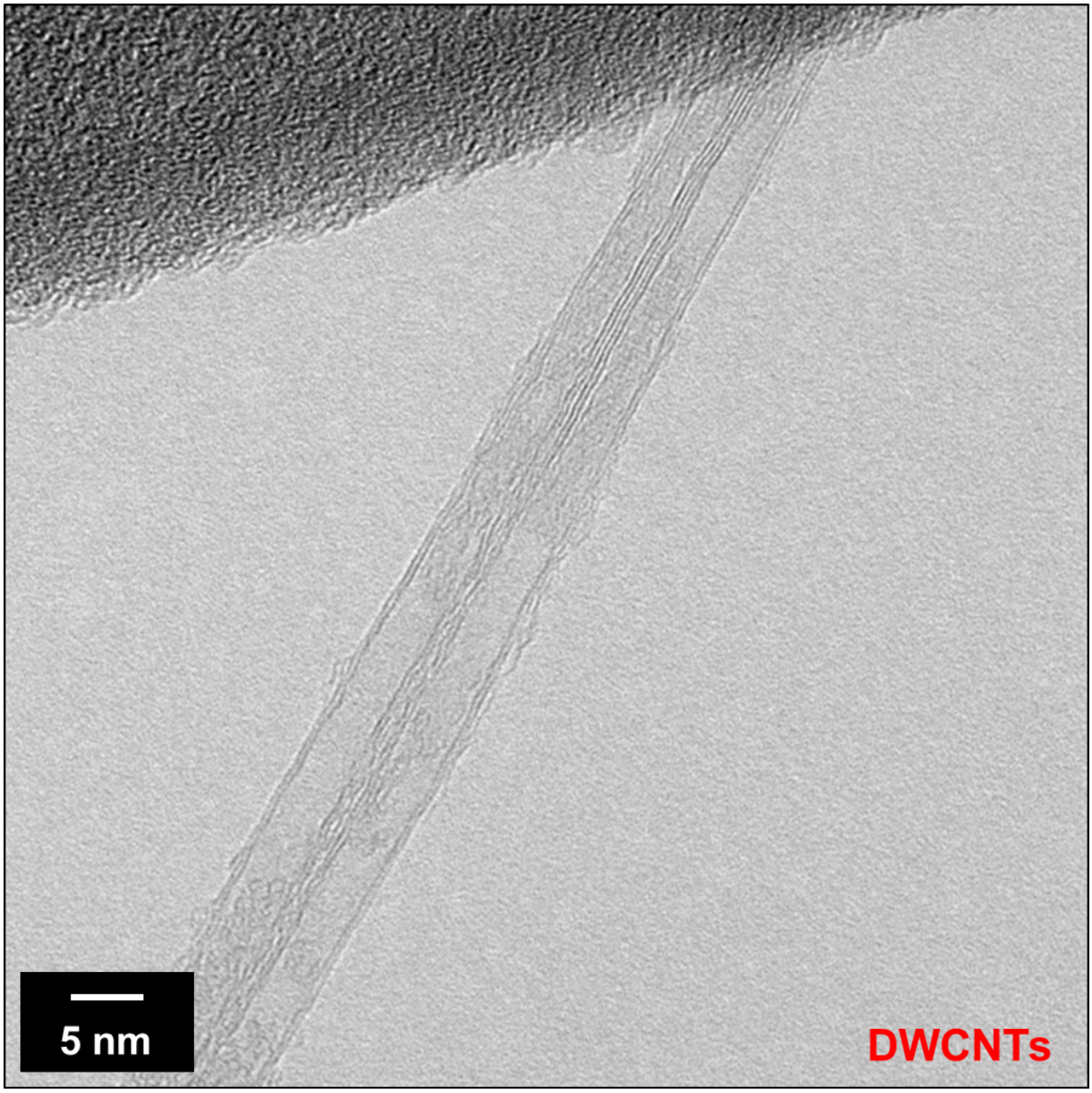

Fig. S1-1 TEM image of DWCNTs synthesized from acetone, ferrocene, and thiophene

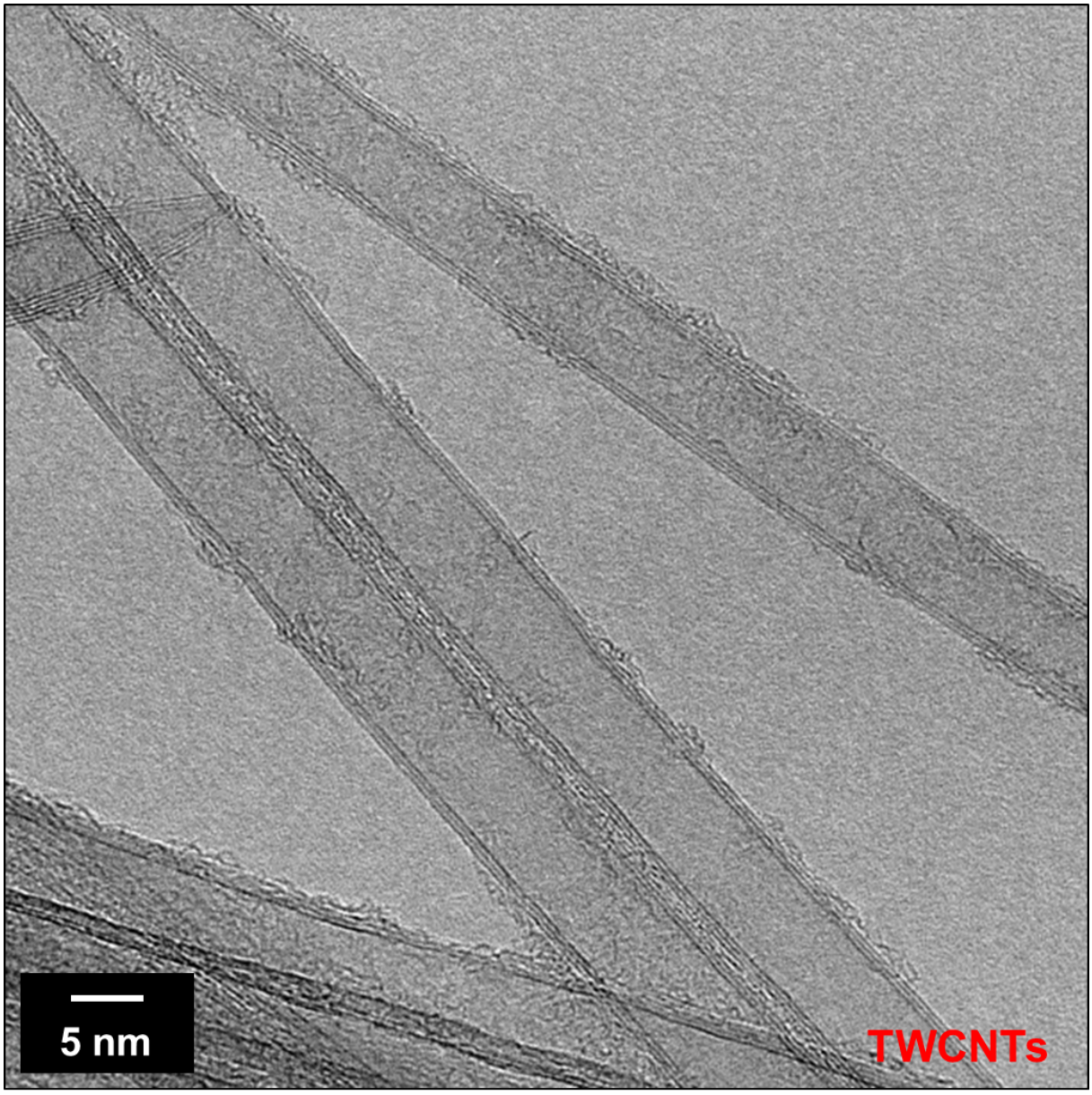

Fig. S1-2 TEM image of TWCNTs synthesized from acetone, ferrocene, and thiophene

Supplementary information 2 (S2) : The strength of synthesized CNT fibers

The strength of a CNT fiber is influenced by many factors: quality (ex, length, purity and crystallinity) of constituent CNTs, alignment of CNTs in a CNT bundle, amount and distribution of voids between CNT bundles, just name a few. These factors can be classified into two categories: material properties and other properties related to the processing conditions. Material properties are of primary concern in this paper because the factors related to the processing conditions are strongly dependent upon processing environments which are very difficult to reproduce in different laboratories of different locations and different times.

The strengths of the CNT fibers obtained in our experiments are shown in Fig. S2-1. The strength of our CNT fibers trends to decrease with the decreasing quality of CNTs (from SWCNTs, DWCNTs to MWCNTs). This trend is in accordance with the results of Raman analysis appeared in Table S1. The $\mathbf{I_G/I_D}$ ratio also decreases with the quality of CNTs.

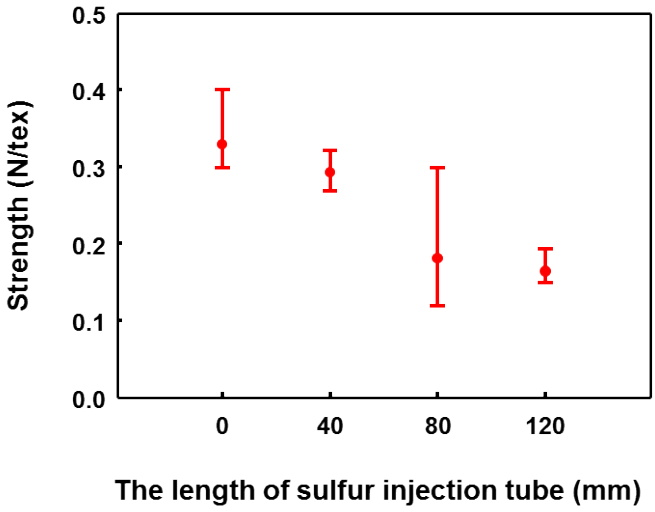

Fig. S2-1 Mechanical properties of CNT fibers

Table. S1 Raman I_G/I_D and mechanical properties of CNT fibers

Promoter	Tube length (mm)	The type of CNTs	I_G/I_D	Average strength (N/tex)
Sulfur	0	SWCNTs	26.9	0.33
	40	SWCNTs+DWCNTs	17.6	0.29
	80	DWCNTs	11.8	0.18
	120	MWCNTs	1.5	0.16

Although the trend is clear, the average strengths of these CNT fibers are not very high. We attribute these not-so-great average strengths to the voids distributed throughout our CNT fibers. Fig. S2-2 shows the surface of our CNT fiber, and it can be immediately noticed that this fiber needs further compaction to be a stronger one. It has been known that compaction increases the strength of a CNT fiber (1). Our own investigation on the compaction of CNT fibers is ongoing, and will be published later.

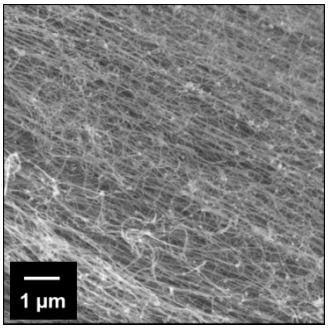

Fig. S2-2 The surface of CNT fiber synthesized from methane, ferrocene, and sulfur

Reference

1) J.N. Wang, X.G. Luo, T.Wu & Y. Chen, *Nature Communications*, 2014, **5**, article number: 3848, doi:10.1038/ncomms4848

Supplementary information 3 (S3) :

The size distribution of catalyst particles and those of synthesized CNTs

The positions of sulfur injection tubes were (a) 0 mm, (b) 40 mm, (c) 80 mm, and (d) 120 mm from the top of the vertical tube reactor. We measured the size of 50~70 catalyst particles of each sample (in the case of 120 mm, 26 catalyst particles were measured), and the size distributions of catalyst particles are given in Fig. S3. Fig. S3-1 shows the size distributions for catalyst particles, which move to the right (which corresponds to the direction of larger catalyst size) and become wider as the length of the sulfur injection tube increases. A similar trend is observed in the diameter distributions of synthesized CNTs, which is shown in Fig. S3-2. The actual size data for catalyst particles and synthesized CNTs are also displayed as histograms.

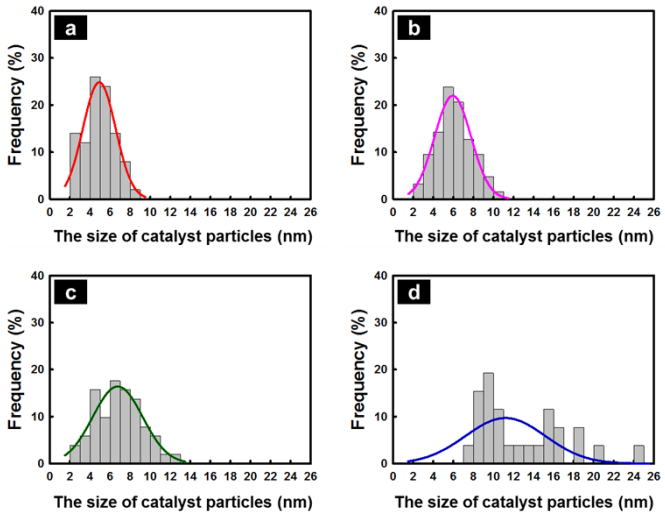

Fig. S3-1 The histograms and distribution curves of the size of catalyst particles.

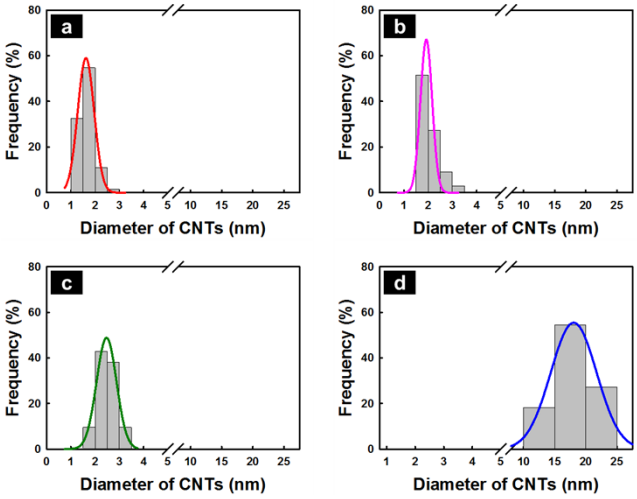

Fig. S3-2 The histograms and distribution curves of diameter of CNTs.