

Optimization of the catalytic chemical vapor deposition synthesis of multi-wall carbon nanotubes on FeCo(Ni)/SiO₂ aerogel catalysts by statistical design of experiments

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The quality of the nanotube product obtained by the 49 catalytic runs (which differ by catalyst composition and/or catalytic deposition conditions) was assessed by analysis of the TEM images of the product of the different experiments.

In order to define a nanotube quality-related parameter, the TEM images were evaluated by a score ranging from 0-3 defined as follows:

<i>TEM score</i>	<i>Quality of the Nanotubes</i>
0	poor nanotube production/quality
1	MWCNTs with different lengths/diameters
2	good quality MWCNTs and small fractions of lower quality nanotubes
3	only good quality MWCNTs without detectable contamination

We have evaluated at least 7 TEM images taken at different spots for each sample using a scale of 0, 1, 2 and 3 quality score, and Figure S.1 reports additional examples of TEM images of samples grouped by score levels. Note that the TEM analysis was performed on the nanotubes as deposited on the catalysts, in order to avoid potential artifacts due to the purification procedures. As a consequence, portions of the nanocomposite catalyst can also be observed in TEM images.

As pointed out in the manuscript, both the quality and the quantity of the carbon deposit varied largely during the 49 experiments.

For example, runs which were given quality score 1 include catalytic runs with different carbon yields (%C ranges from 49.4 and 43.5 to 652.8 for runs #14, 36, 46). Likewise, runs which were scored 2 include catalytic runs with %C 242.2 for run #6 and 1.5 for run #3.

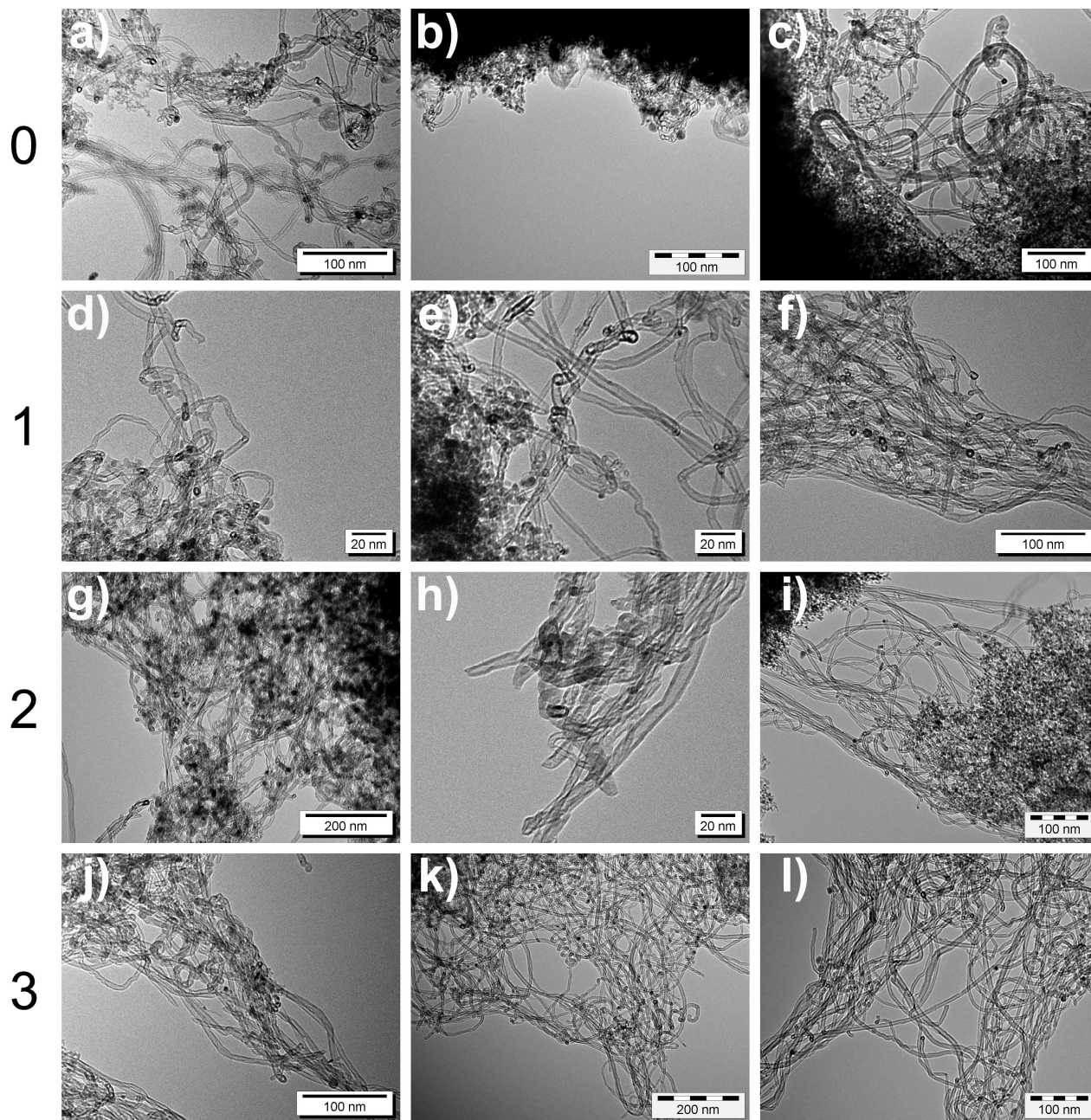


Figure S.1 Further examples of representative TEM images of carbon nanotube products evaluated by quality score: images a), (run #39), b) (run #37) and c) (run #45) were scored 0; images d) (run #14), e) (run #36) and f) (run #46) were scored 1; images g) (run #6), h) (run #11) and i) (run #3) were scored 2; images j) (run #44), k) (run #42) and l) (run #30) were scored 3.

Figure S.2 shows additional TEM images of the product obtained by run #30, which based on the observed presence of long carbon nanotubes with a homogeneous diameter without any other carbonaceous deposit was scored of quality 3. It should be pointed out that the statistical analysis of all the collected data suggest that the optimized response is obtained for conditions very similar to the ones adopted for the synthesis and catalytic parameters adopted for run #30. In particular, the TEM quality score predicted for the optimum set of conditions is $TEM=2.66$, similar to the score value attributed to run #30 based on the analysis of the obtained images.

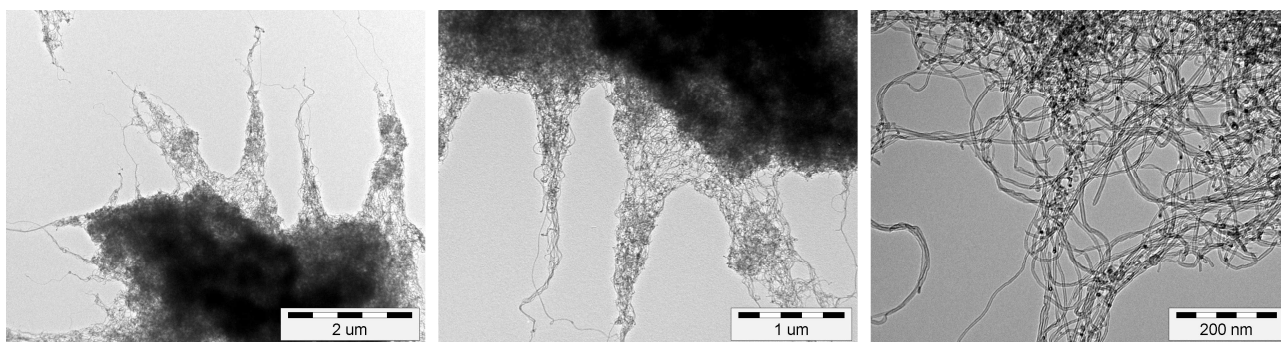


Figure S.2 TEM images of run #30, which gave production of carbon nanotubes of high quality and high yield and approximates the optimum set of conditions obtained by data analysis.