

## Supporting Information

### Molecular Encapsulation from the Liquid Phase and Graphene Nanoribbon Growth in Carbon Nanotubes

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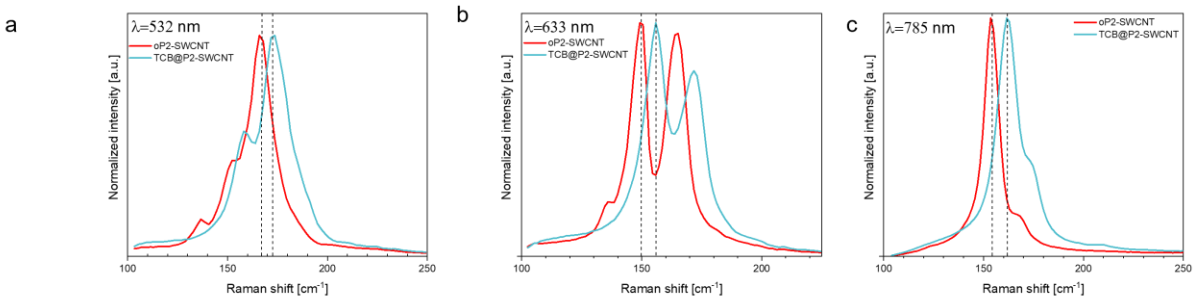


Figure S1. Raman spectra of empty and TCB-filled nanotubes in the RBM region measured with different laser excitation.

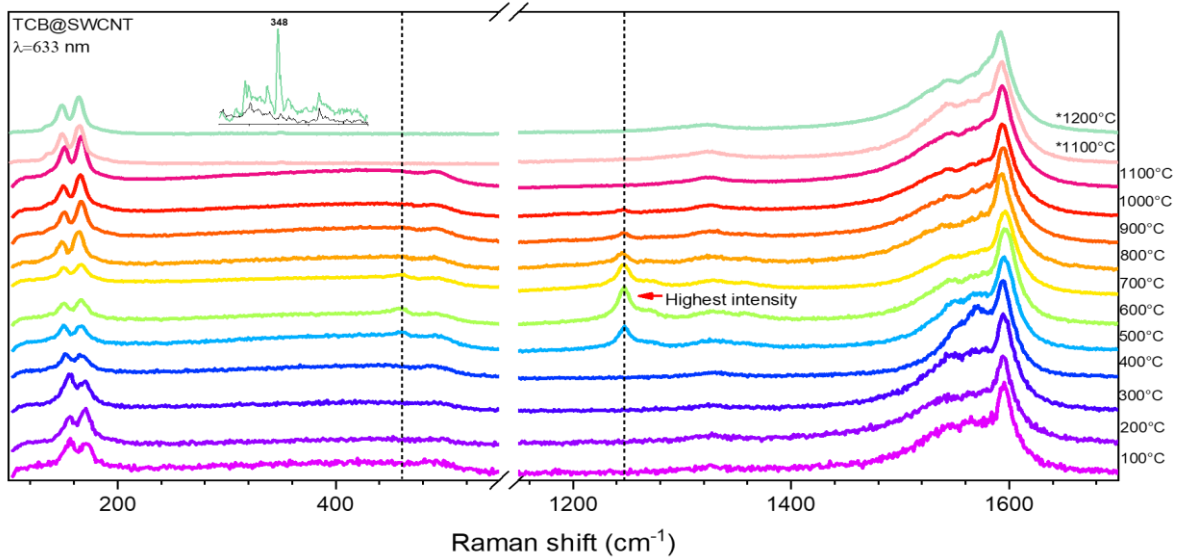
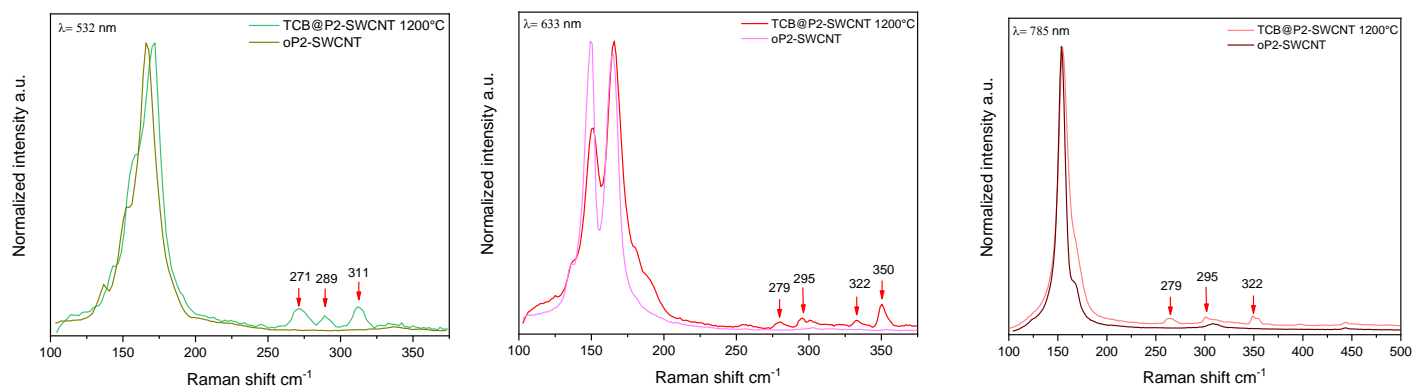


Figure S2 Raman spectra following the ribbon formation, consecutively annealing from 100 °C to 1200 °C in 100 °C increments. The highest intensity peaks of the 6-AGNR, indicated by dashed lines, appeared at 500 °C and 600 °C. The spectra was measured through the quartz tube, for the temperatures made with \* the quartz tube was opened and the spectra was measured straight on the sample. The inset shows the double wall picks formed at 1200 °C (green) vs the SWCNT with open caps (black).



**Figure S3.** The RBM region of double-walled carbon nanotubes formed at 1200 °C after annealing the TCB@P2 SWCNT 600 °C sample.

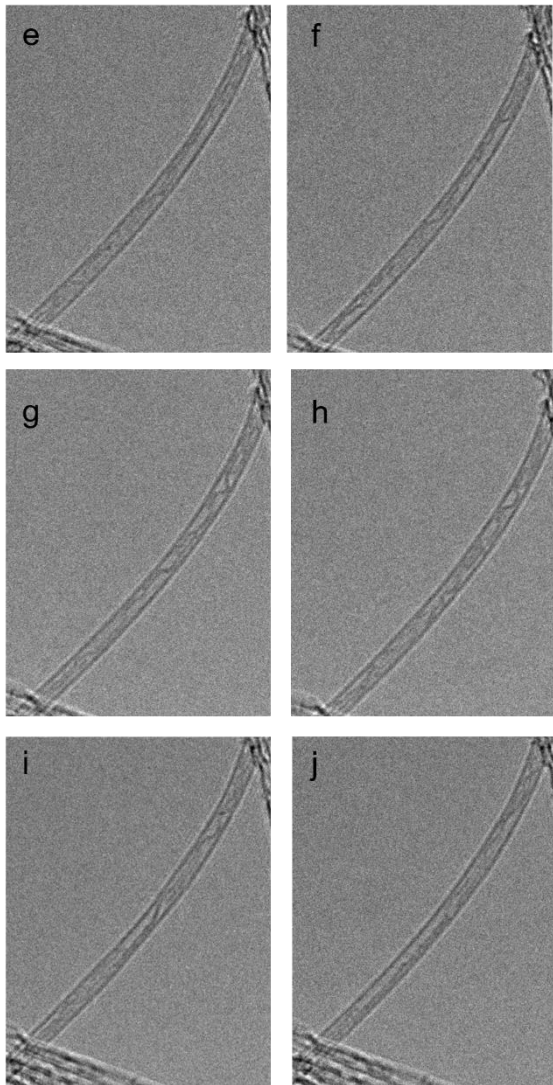
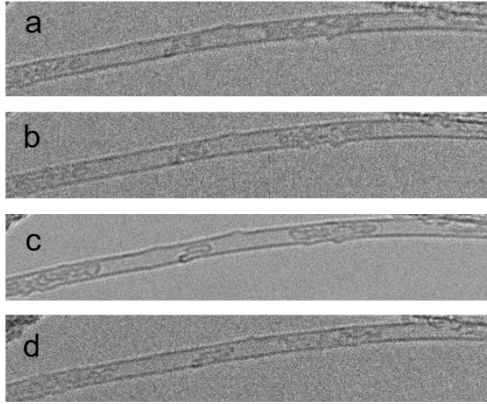
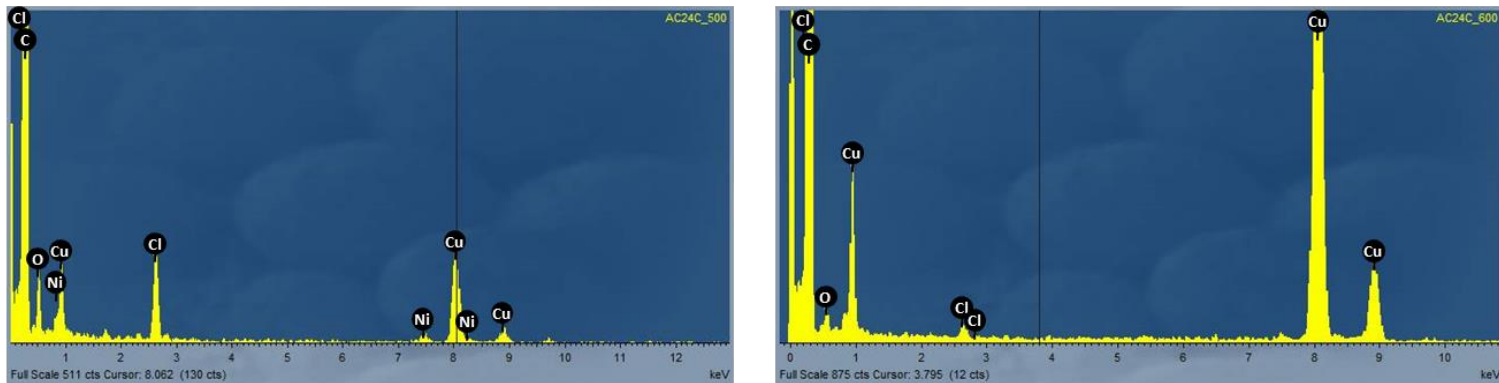
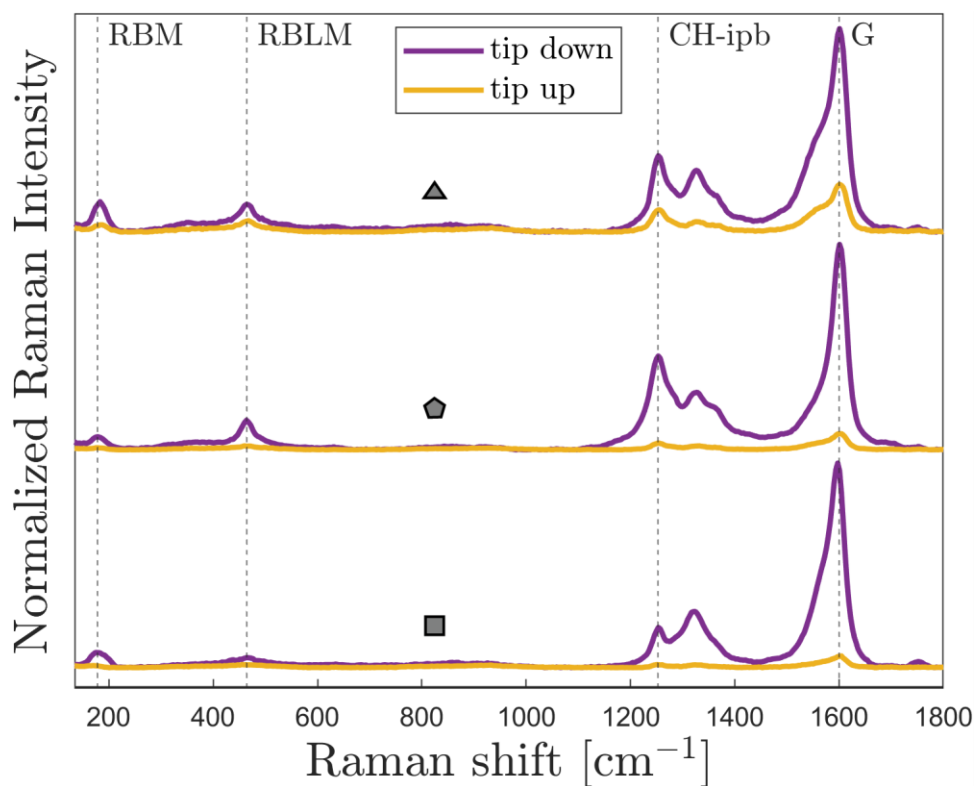


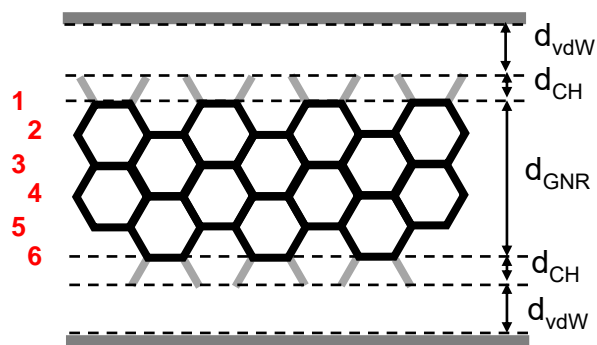
Figure S4. Time series TEM images on nanoribbons formed inside the nanotubes at 500C° (a,b,c,d) and 600C° (e,f,g,h,i,j) annealing temperature.



**Figure S5.** EDS spectra of the ribbons formed at a) 500°C and b) 600° C.



**Figure S6.** Raman spectra of GNR@P2-SWCNT recorded at the locations marked in Fig. 5(d) with (tip down) and without (tip up) the presence of the TERS probe. All spectra are normalized to the G peak height of the TERS measurements and vertically offset for better visibility.



**Figure S7.** Calculation of the width of armchair (AGNR) nanoribbons. The model is based on simple geometric considerations, using  $a_{CC}=0.142$ ,  $a_{CH}=0.11$  as carbon-carbon and carbon-hydrogen bond length respectively. The van der Waals distance between the hydrogen and the carbon nanotube wall was calculated based on the van der Waals radii of the atoms ( $d_{vdW}=0.29$  nm).

N	AGNR	ZGNR	AGNR+H	ZGNR+H	AGNR+H+vdW	ZGNR+H+vdW
2	0.123	0.284	0.314	0.504	0.894	1.084
3	0.246	0.497	0.436	0.717	1.016	1.297
4	0.369	0.71	0.559	0.93	1.139	1.51
5	0.492	0.923	0.682	1.143	1.262	1.723
6	0.615	1.136	0.805	1.356	1.385	1.936
7	0.738	1.349	0.928	1.569	1.508	2.149
8	0.861	1.562	1.051	1.782	1.631	2.362

**Table S1** Width of the nanoribbons with different N-numbers. Column AGNR and ZGNR contains the width of the bare carbon network. AGNR+H and ZGNR+H includes the terminating hydrogens. The last two columns (AGNR+H+vdW, ZGNR+H+vdW) incorporates the van der Waals distance between the carbon nanotube wall and the terminating hydrogens. Green background indicates the ribbons which can be fitted inside the CNTs considering the threshold for ribbon formation.