Supporting Information

Cutting Single-walled Carbon Nanotubes with a 'CO_2 Blade'

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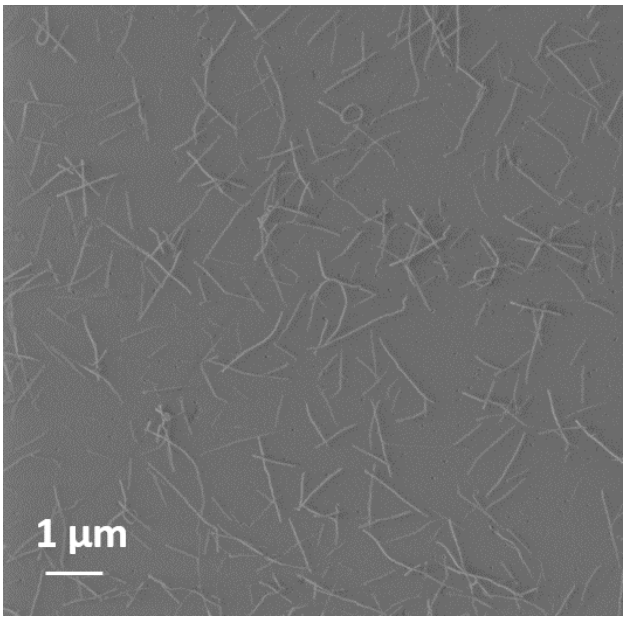

Figure S1. SEM image of the SWCNTs collected from *Reactor II* without adding CO_2, which presents similar mean length of ≈1.0 μm as that of as-synthesized SWCNTs.

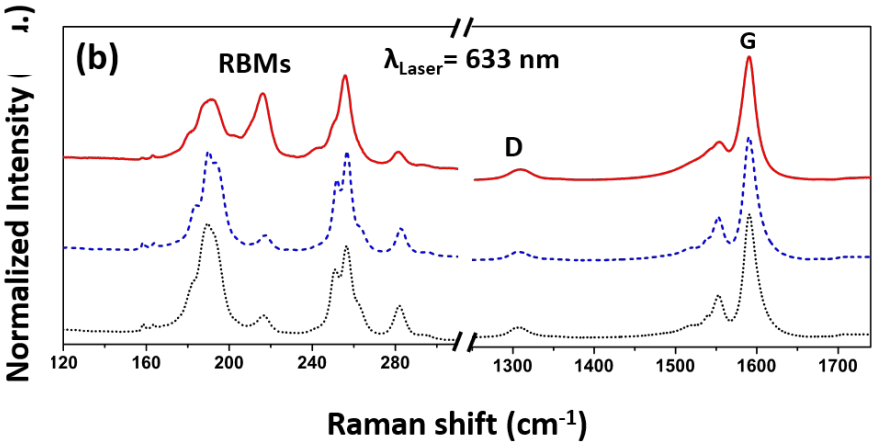

Figure S2. Raman spectra of as-synthesized SWCNTs collected from *Reactor I* (black dots), cut SWCNTs deposited from *Reactor II* at CO_2 concentration of 6% (blue dashes) and 43% (red solid lines), respectively, at λ_{Laser} = 633 nm. The intensities of D bands are

comparable for the as-synthesized and cut-SWCNT samples, indicating that the quality of the shortened SWCNTs remains high.

Grand Canonical Monte Carlo (GCMC) Calculation

We use computer simulations to gain an insight at the atomic scale into possible scenarios of the cutting process. In the same spirit as using GCMC simulations for growing nanotubes on a catalyst nanoparticle by incorporating carbon atoms in the structure at given temperature (T) and chemical potential (μ_C) [1], we use the GCMC algorithm to remove carbon atoms from an existing structure. The chemical reaction that involves CO_2 to remove the less stable carbon (C) atoms through a reverse Boudouard reaction, translates in our GCMC algorithm into a change of μ_C. At 1000 K, we thus shift from values previously used to favor nanotube growth, typically around -6 eV/ C atom, to more negative ones, around -9 eV/C atoms, that induce a controlled etching of C atoms. We study a configuration where a C_{60} fullerene is attached to a (14,0) tube, removing the 'overlapping' C atoms and relaxing the full system, in such a way that all but three carbon atoms are 3-fold coordinated, for total of 498 atoms. We then performed GCMC calculations at the conditions of T=1000 K and μ_C = -8.90 eV / atom. Snapshots of atomic configurations resulting from the GCMC etching of C atoms are depicted in Figure S3. A full video is also provided as supplementary material.

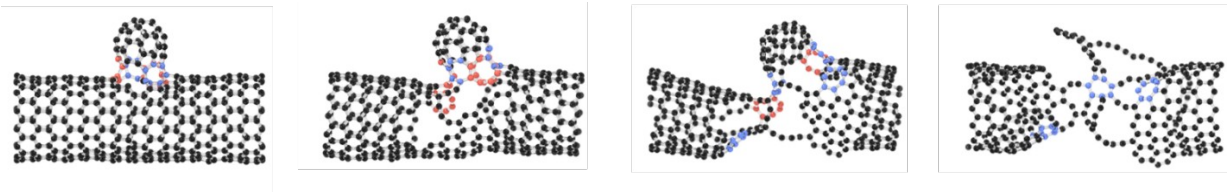

Figure S3. Snapshots of atomic configurations resulting from the GCMC etching of carbon atoms from a starting configuration formed by attaching an open C_{60} fullerene cap to a (14,0) tube. All carbon atoms are in black, except those belonging to heptagons (blue) or octagons (red). Starting from the configuration, defects are formed in the tube structure, leading to an almost complete cut and partial healing of the open lips. Note that left and right sides of the tube are connected through so-called periodic boundary conditions.

[1] Amara, H.; Bichara, C.; Ducastelle, F., Understanding the nucleation mechanisms of carbon nanotubes in catalytic chemical vapor deposition. *Physical review letters* **2008,** *100* (5), 056105.