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Restoration of the Genuine Electronic Properties of Functionalized Single-Walled Carbon Nanotubes

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FT-IR spectrum and peak assignments of functionalized SWNTs

Figure S1 shows the background-subtracted peak intensities of the hydroxyphenyl groups attached to the sidewalls of SWNTs at various annealing temperatures, i.e., O-H stretching (~3360 cm^{-1}), C-H stretching (~2920 cm^{-1}), and C=C stretching (~1540 cm^{-1}) as well as the C-H bending (~1200 cm^{-1}) mode. The change in the peak intensity of each functional group upon annealing, shown in Figure 2 of original manuscript, were normalized to the intensity of the fully functionalized SWNT sample of Figure S1.

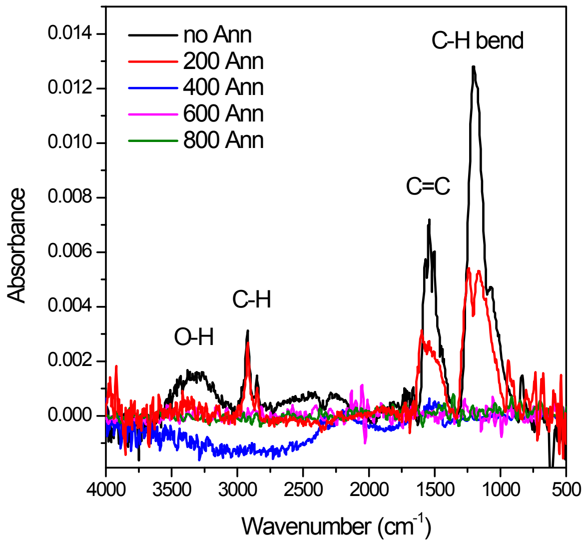

Figure S1. Changes in the FT-IR peak intensities of functional groups of hydroxyphenyl groups attached on the sidewalls of SWNTs upon annealing at various temperatures.

Restoration of the genuine electronic properties of carboxyphenyl functionalized Single-Walled Carbon Nanotubes

We also investigated that the annealing treatment of this study is a universal technique in effectively removing other kinds of functional groups, rather than hydroxyphenyl functional groups used in this study. We used carboxyphenyl group (carboxybenzene diazonium reagent, $COOH\text{-}C_6H_4\text{-}N_2^+BF_4^-$, was used to attach carboxyphenyl group onto SWNT surfaces) as an alternative functional group other than hydroxylphenyl group, to functionalize SWNT and annealed the functionalized SWNT at various temperatures. Figure S2(a) shows the changes in the electrical properties of carboxyphenyl functionalized SWNT. As can be seen in Figure

S2(a), the sheet resistance of carboxyphenyl functionalized SWNT is recovered to the level of pristine SWNT (without functionalization) upon annealing at above 400°C, similar to the case of hydroxyphenyl functionalized SWNT. Figure S2(b) shows the Raman spectra of carboxyphenyl functionalized SWNT without annealing, functionalized SWNT anneaded at 400°C and 600°C, respectively. We can clearly see that the carboxyphenyl functional groups are removed above 400°C, as evidenced by the decrease of disorder mode (~1300 cm-1), similar to the case of hydoxyphenyl functionalized SWNT of the original manuscript. Based on the Raman and electrical property measurement experiments, we concluded that the annealing condition (annealing at above 400°C is necessary for removal of functional groups) used in this study can be applied for removing not only hydroxyphenyl groups but also other functional groups from SWNT surfaces.

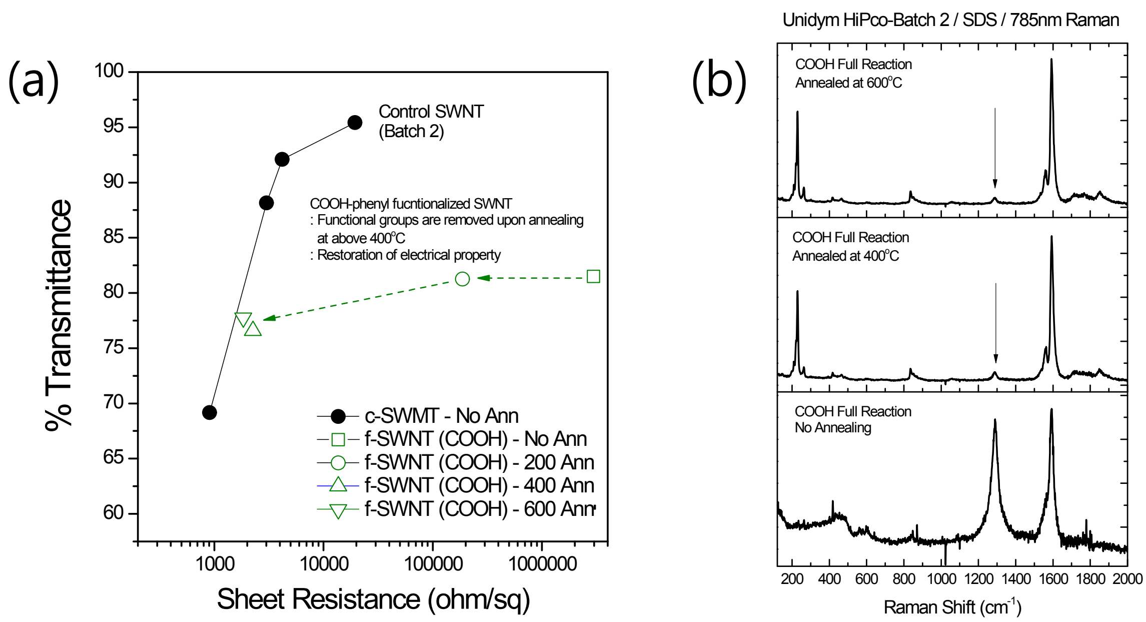

Figure S2. (a) The sheet resistance versus transmittance of unfunctionalized (c-SWNT, filled symbol) and carboxyphenyl functionalized SWNTs (f-SWNT, hollow symbol) films before and after annealing at various temperatures. (b) Raman spectra of unfunctionalized (c-SWNT) and carboxyphenyl functionalized SWNTs (f-SWNT) and those of functionalized SWNTs annealed at various temperatures.

Reversibility of functionalization and annealing process

To investigate that this annealing process can be applied to the re-functionalized SWNTs after first annealing process, we re-functionalized the carbon nanotubes after annealing treatment and showed the results of second annealing process in Figure S3. As can be seen in Figure S3, re-functionalization and re-annealing process can be regarded as reversible, even though there is a slight difference in the sheet resistance values between the first and second annealing treatment at each annealing temperature.

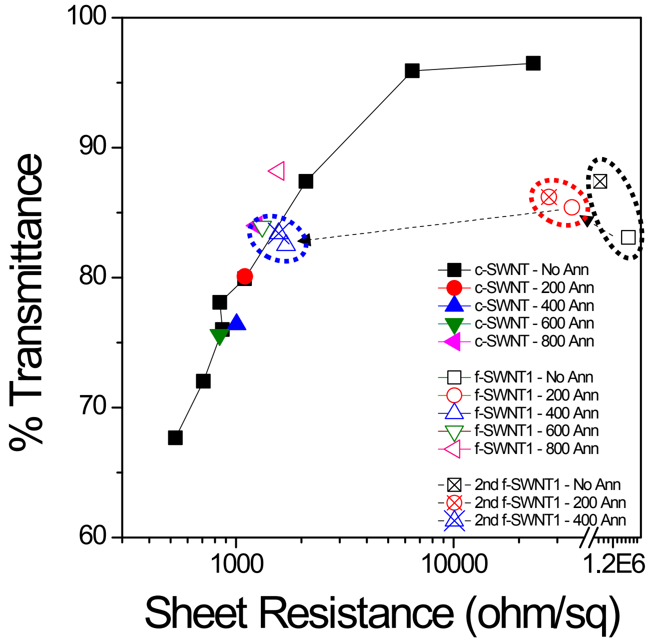

Figure S3. The sheet resistance versus transmittance of unfunctionalized (c-SWNTs, filled symbol) and first- and second-functionalized SWNTs (f-SWNT, hollow symbol) films before and after annealing at various temperatures.

UV-vis-nIR transmittance spectra of annealed metallic SWNTs

Figure S2 shows the UV-vis-nIR transmittance spectra of SWNT mixture film and separated metallic SWNT film, annealed at 800°C. Annealing at at 800°C removes all of the functional groups from metallic SWNTs, and recovers typical second- (E_{22}^{SC}, 600 ~ 800 nm) order optical transitions of semiconducting SWNTs and first-order (E_{11}^{M}, 440 ~ 645 nm) optical transitions of metallic SWNTs. Separated and annealed metallic SWNT film shows increased intensity for E_{11}^{M} and decreased intensity for E_{22}^{SC}, compared to SWNT mixtures, indicating that the high purity metallic SWNTs are obtained. This sample was used for electrical measurement shown in Figure 6 of the original manuscript.

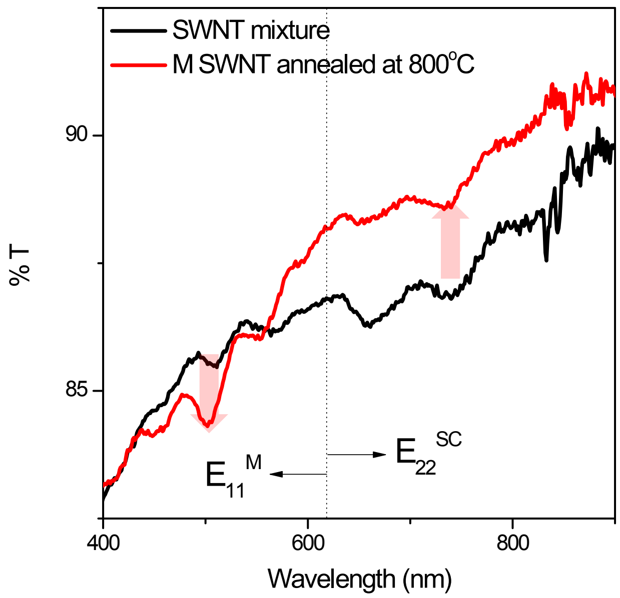

Figure S4. UV-vis-nIR transmittance spectra of SWNT mixtures and annealed metallic SWNTs.