Supplementary Information

Carboxylation of thin graphitic sheets is faster than that of carbon nanohorns

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Supplementary Information 1. Structure of CNH-TGS aggregate did not show apparent changes after the H_2O_2 treatment. TEM image of CNH-GLS aggregate treated with aqueous solution of H_2O_2 for 7 days (Figure SI-1) shows CNHs and GLSs without obvious defects.

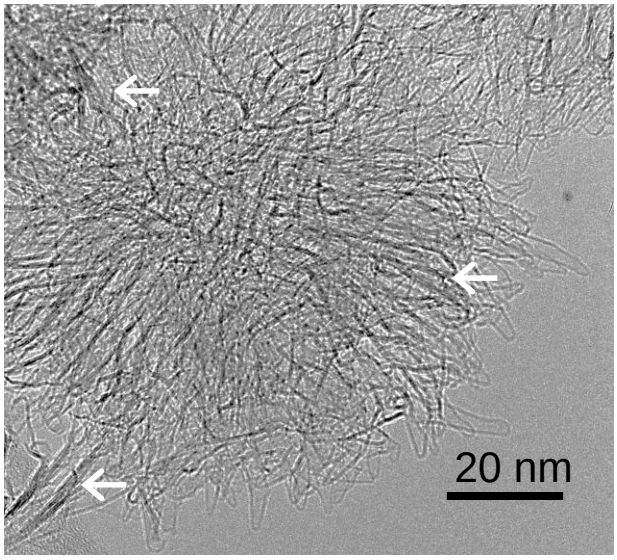

Figure SI-1. TEM image of CNH-GLS aggregate treated with aqueous solution of H_2O_2 for 7 days. GLSs are indicated with arrows.

Supplementary Information 2. Z-contrast image of CNH-GLS with Pt-ammine complex reacted with carboxyl groups. Figure SI-2 shows that the Pt complex clusters did not exist on CNHs.

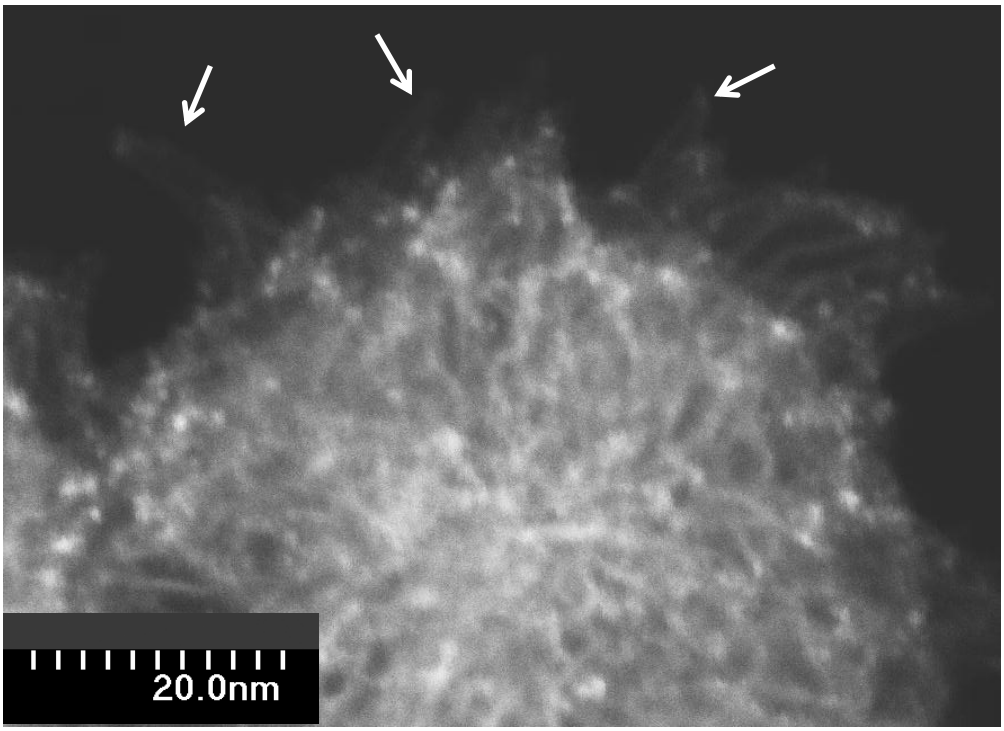

Figure SI-2. Z-contrast image of CNH-GLS with Pt-ammine complex reacted with carboxyl groups. Arrows indicate CNHs. Bright spots are Pt-complex clusters. Bright lines correspond to the side views of GLS or CNH walls.

Supplementary Information 3. EDX analysis was performed on individual CNH-GLS aggregates treated with Pt ammine complex. The results showed that the aggregate contained Pt and typical data are shown in Figure SI-3.

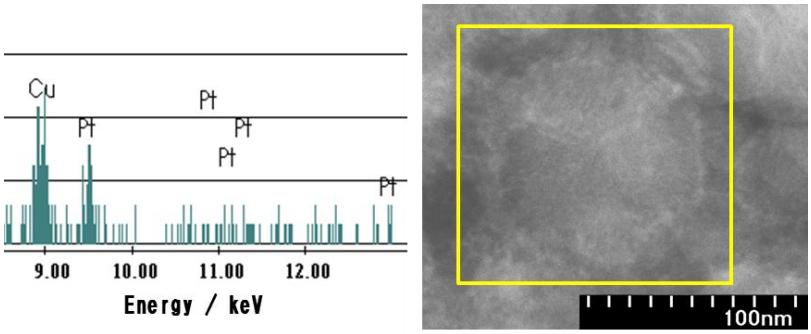

Figure SI-3. Energy dispersive spectrum (left panel) of one aggregate measured for one aggregate in a yellow square region of a Z-contrast image (right panel). A peak corresponding to Cu in EDX spectrum was originated from a Cu grid sample holder.

Supplementary Information 4. Infrared absorption (IR) spectra were measured to confirm that Pt ammine complex existed in CNH-GLS treated with Pt complex.

In the measurements, the specimens were dispersed in ethanol (2 mg/mL) and sprayed on the ZnSe single crystal plate (diameter 2 cm, thickness 5 mm). Spectra were measured with Perkin Elmer Spectra One.

The IR spectra of CNH-GLS treated with Pt ammine complex showed the absorption peaks characteristic of NH_3 moieties of the Pt ammine complex (Figure SI-4, purple line) [1], while the CNH-GLS before the treatment with Pt ammine complex did not show these peaks. This supports the result that the Pt ammine complex reacted with the carboxyl groups generated at GLS edges treated with H_2O_2 for 1h.

Probably because the number of carboxyl groups of CNH-GLS treated with H_2O_2 for 1h was not enough, the peaks corresponding to C=O of carboxyl groups in 1600-1800 cm^{-1} range were not clearly visible in IR spectra (Figure SI-4, blue line). The peaks at 1180 and 1560 cm^{-1} are intrinsic ones of CNH-GLS (Figure SI-4, red lines).

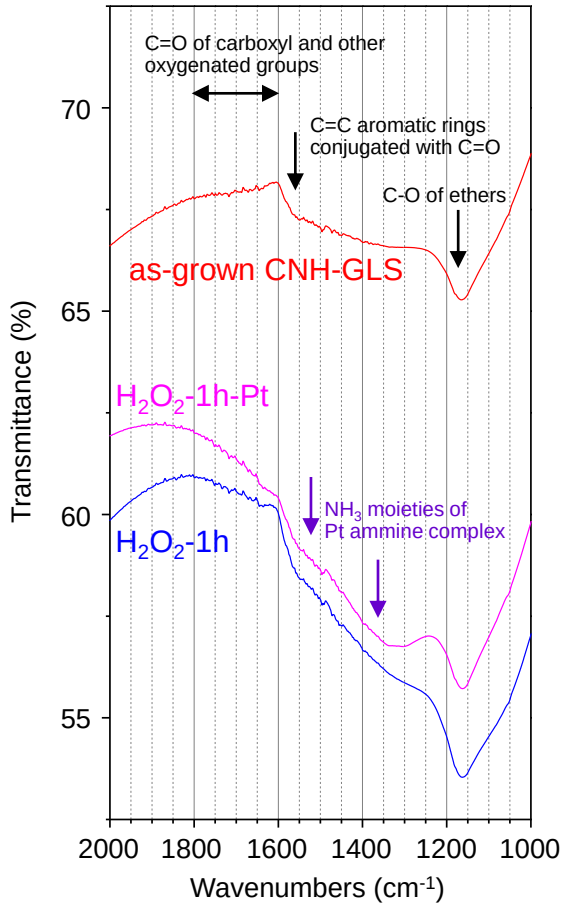

Figure SI-4 Infrared absorption spectra of as-grown CNH-GLS (red line), after treatment with H_2O_2 for 1 hour (blue line), and further treated with Pt ammine complex (magenta line).

Ref. [1] R. Yuge, M. Zhang, M. Tomonari, T. Yoshitake, S. Iijima and M. Yudasaka, *ACSNano*, 2008, **2**, 1865-1870.

Supplementary Information 5. The Z-contrast image in Figure 4 shows about 4-5 bright spots of Pt-compound clusters (sizes: about 1 nm) in the 10-nm length of edges of GLSs. The number of carboxyl groups in 10-nm length TGS edges (zigzag or armchair type) is about 40. Here, it is assumed that the C-C distances were 0.142 nm taken from graphite C-C length and, as shown in the text, 50% of carbon atoms at the edges were of carboxyl groups. Then it turns out that there were 8-10 Pt-compounds in each 1-nm size cluster. Or each Pt-compound had the size of about 0.4 nm, which seems to be reasonable.