Supplementary Information for

Ultrafast, Controllable Synthesis of Sub-Nano Metallic Clusters through Defect Engineering

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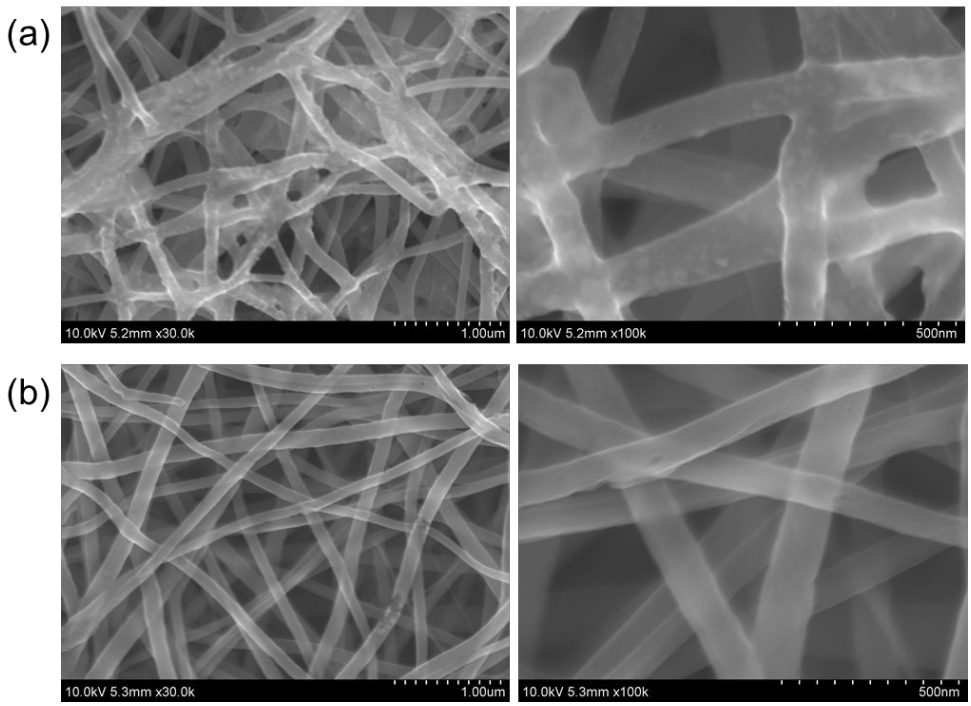

Fig. S1. SEM images of (a) 5 μmol/cm^2 and (b) 0.5 μmol/cm^2 Pt precursor salt loaded CA-CNFs.

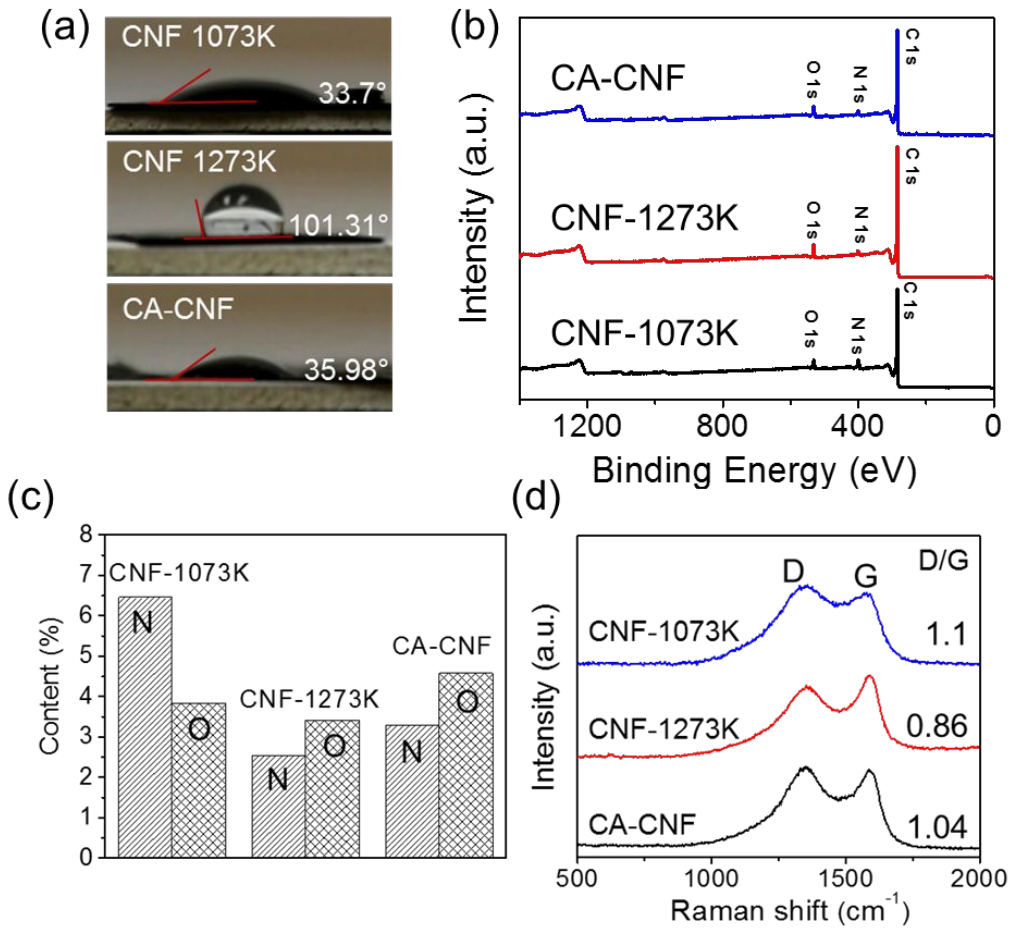

Fig. S2. (a) Wetting angle measurement, (b) XPS spectra, (c) N and O content from XPS spectra and. (d) The Raman spectra of CNF-1073K, CNF-1273K, and CA-CNF. The numbers indicate D/G peak ratio.

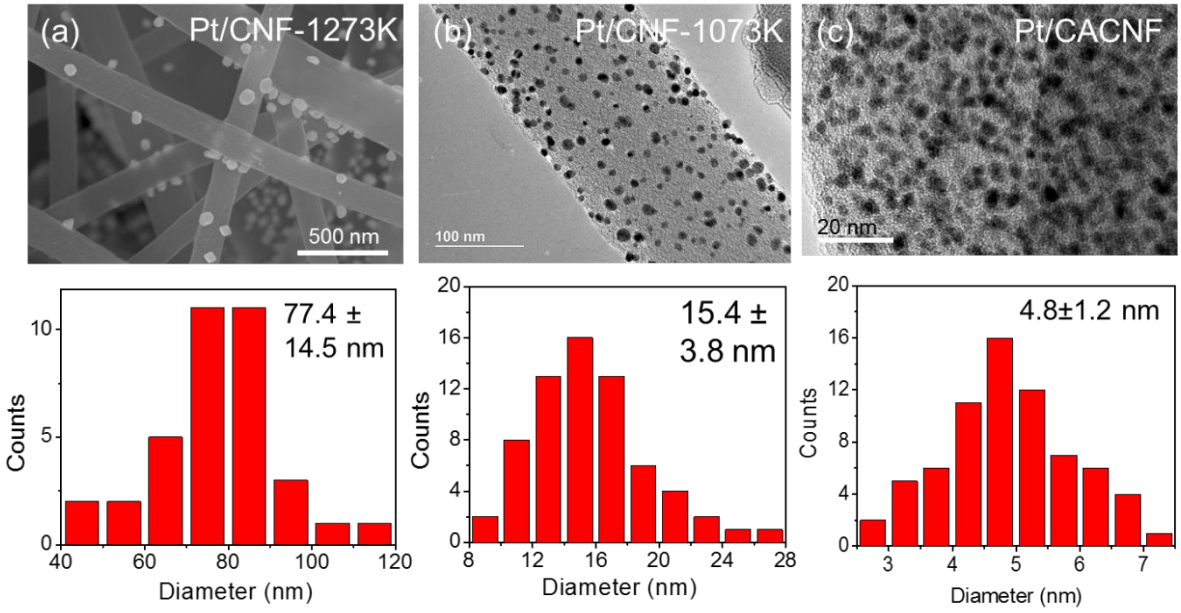

Fig. S3. Pt dispersion on different CNF substrates and corresponding size distribution (synthesized using the same precursor loading (5 μmol/cm^2) and shock process): (a) CNF-1273K, (b) CNF-1073K, and (c) CA-CNF.

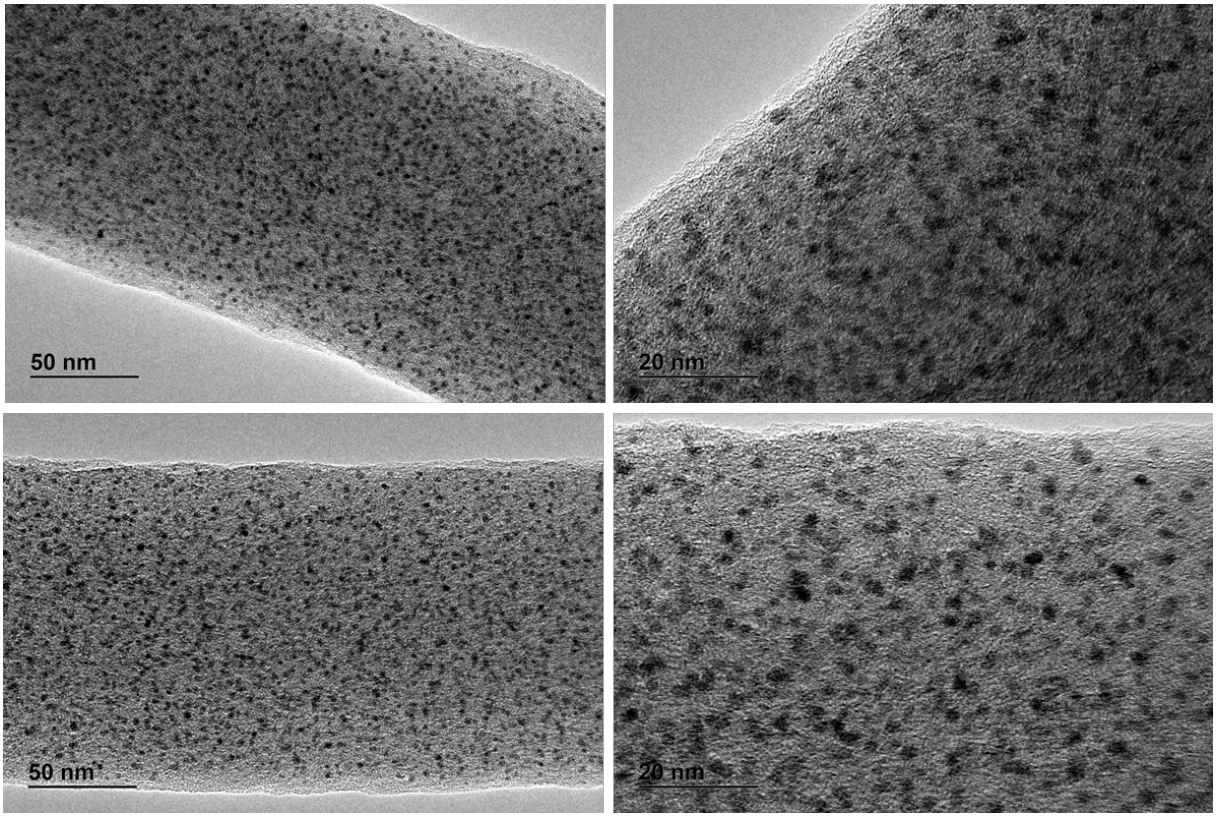

Fig. S4. TEM images of Pt NCs on CA-CNFs (0.5 μmol/cm^2 precursor loading).

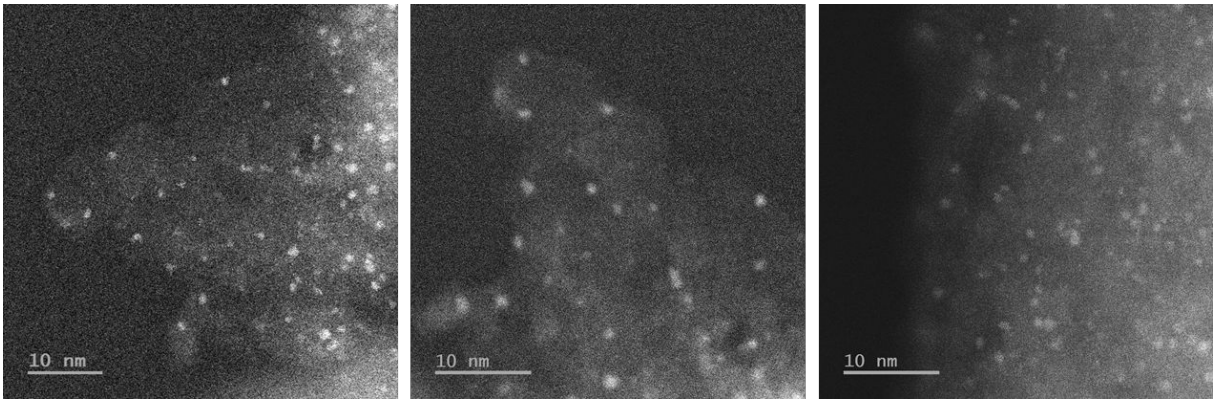

Fig. S5. HAADF images of Pt NCs on CA-CNFs with a lower precursor loading (0.1 μmol/cm^2).

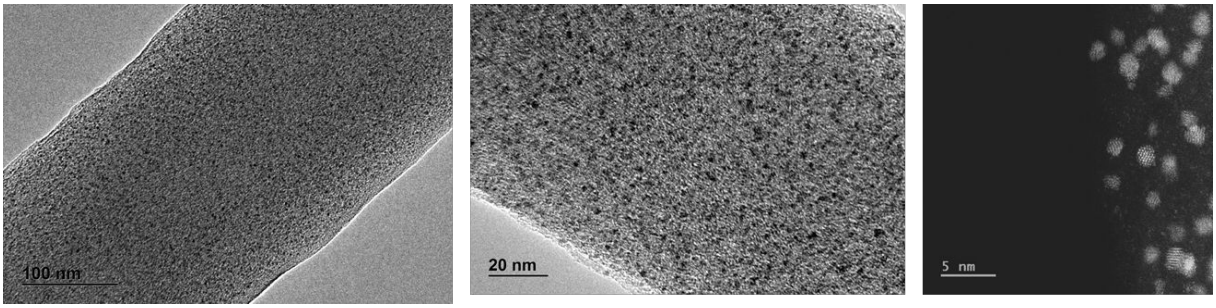

Fig. S6. TEM and HAADF images of Pt NCs on CA-CNFs after CO oxidation, showing well dispersed nanoclusters without obvious aggregation. (0.5 μmol/cm^2) The CO measurement was test from 50°C to 275°C with a 25°C step interval and a heating rate of 5°C/min. The sample was equilibrated for at least 30 minutes before measurement and each measurement take ~ 10 minutes.

Table S1. Size and dispersion density of NCs from our work compared to the literature.

Size (nm)	Density (#/nm^2)	Ref
0.8 - 1.7	~0.1-0.25	Our work
1.6 - 3	~0.1-0.2	*Science* **2018**, *362*, 560-564.
0.94 - 1.4	~0.05-0.2	*Science* **2017**, *358*, 1427–1430.
~1 - 2	<0.01	*Sci. Adv.* **2017**, *3*, e1700101.
1.4 - 2.6	~0.1-0.2	*Angew. Chemie* **2017**, *56*, 2709–2713.
3.76	~0.04	*Int. J. Hydrogen Energy* **2016**, *41*, 8839–8846.
1.7-1.9	~0.01	*Catalysts* **2015**, *5*, 606–633.
0.8	~0.1	*Angew. Chemie - Int. Ed.* **2015**, *54*, 9810–9815.
1.6	~0.1	*Angew. Chemie* **2014**, *126*, 254–258.