

Electronic Supplementary Information for the Manuscript

**Self-Assembly Preparation of Gold Nanoparticles-TiO₂
Nanotube Arrays Binary Hybrid Nanocomposites for Photocatalytic
Applications**

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Synthesis of GNP@DDT nanoparticles

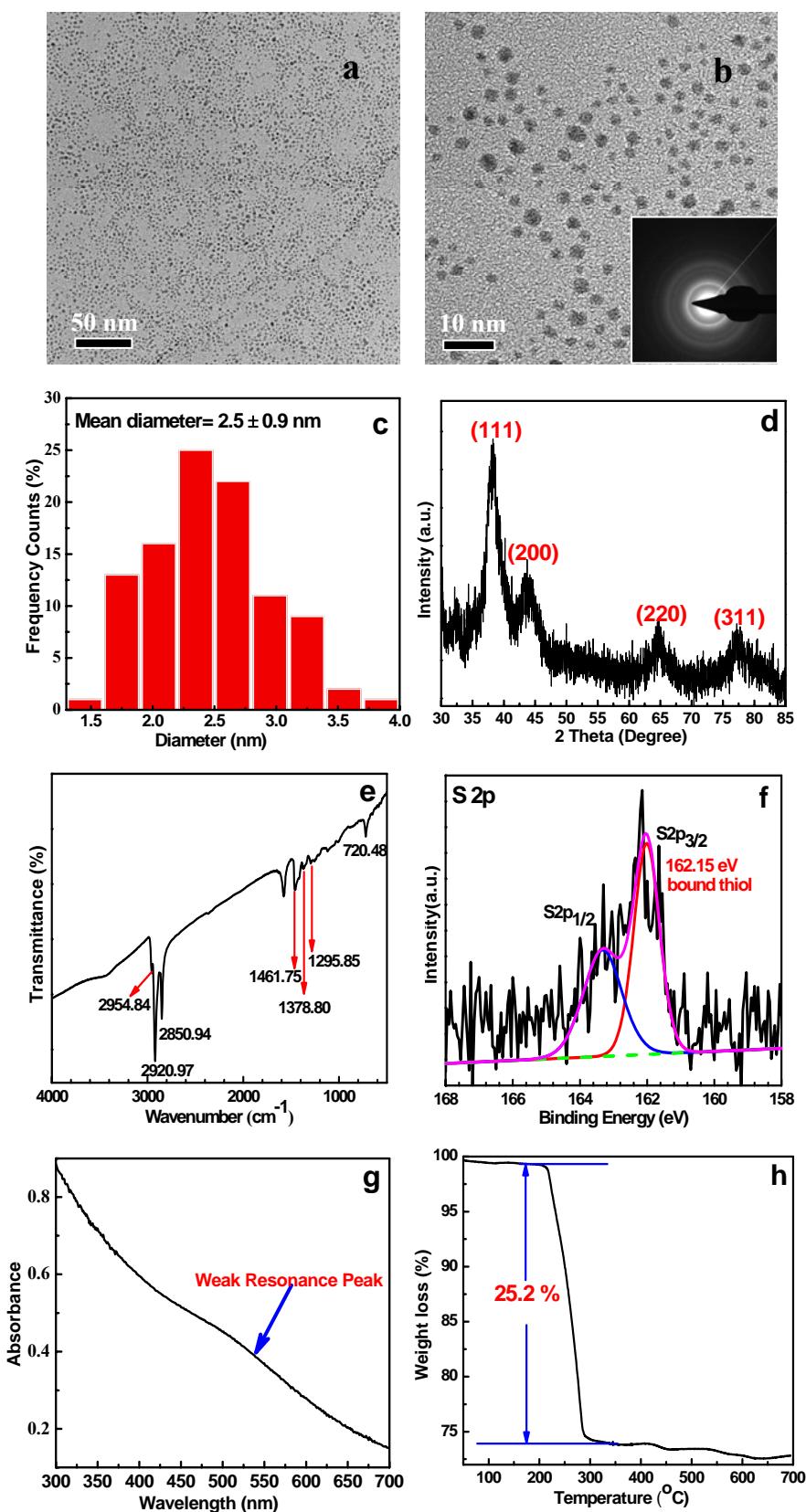


Figure S1. Pre-synthesized GNP@DDT nanoparticles. (a) TEM and (b) HRTEM images; (c) mean diameter histogram (counting to tally of 100 particle s) and (d) XRD pattern of GNP@DDT ; the inset image in (b) showing the repr esentative selected area electron d iffraction (SAED) pattern

for GNP@DDT; (e) FTIR spectrum (CH_3 : 2954.84 cm^{-1} , 1378.80 cm^{-1} , C-C: 1295.85 cm^{-1} , CH_2 : 2920 cm^{-1} , 2850.94 cm^{-1} , 1461.75 cm^{-1} , 720.48 cm^{-1}); (f) High-resolution XPS spectrum of sulfur in GNP@DDT (S 2p, free thiol group -SH: 164.00 eV , bound thiol group -S-Au: 162.15 eV); (g) UV-vis absorption spectrum and (h) TG result of the as-prepared GNP@DDT.

Table S1. Specific chemical bond species *versus* binding energy (BE) in terms of blank TNT As and GNP/TNTAs nanocomposites.

Element	TNTAs (eV)	GNP/TNTAs (eV)	Chemical bond species
C 1s A	284.59	284.61	C-C/C-H
C 1s B	286.27	286.26	C-OH/C-O-C
C 1s C	288.53	288.90	Carboxylate (CO_2 adsorption/MPA)
O 1s A	---	529.00	Oxygen vacancies/Low-energy lattice oxygen
O 1s B	529.82	530.16	Lattice oxygen
O 1s C	531.39	531.08	Ti-OH
O 1s D	532.28	532.09	C-OH/C-O-C
O 1s E	---	533.35	Oxygen species in H_2O
Ti 2p _{3/2}	458.55	458.75	Anatase (+4)
Ti 2p _{1/2}	464.25	464.55	Anatase (+4)
Au 4f _{7/2}	---	83.90	Metallic Au (0)
Au 4f _{5/2}	---	87.45	Metallic Au (0)
S 2p _{3/2}	163.92	162.08	-SH/-S-Au

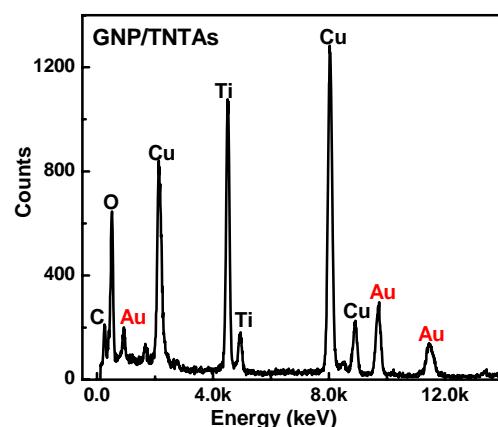


Figure S2. EDX spectrum of GNP/TNTAs

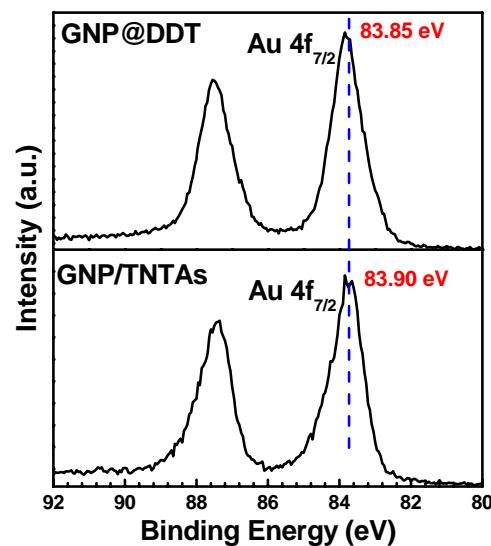


Figure S3. High-resolution Au 4f spectra of GNP@DDT and GNP/TNTAs.

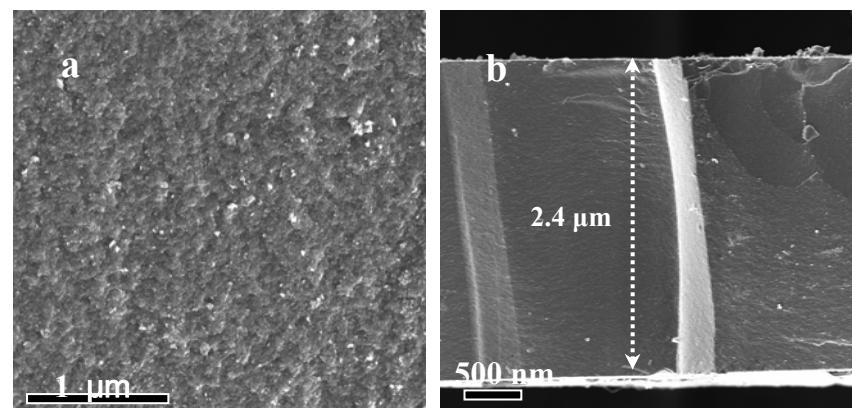


Figure S4. (a) Typical panoramic and (b) cross-sectional FE-SEM images of P25 particulate film.

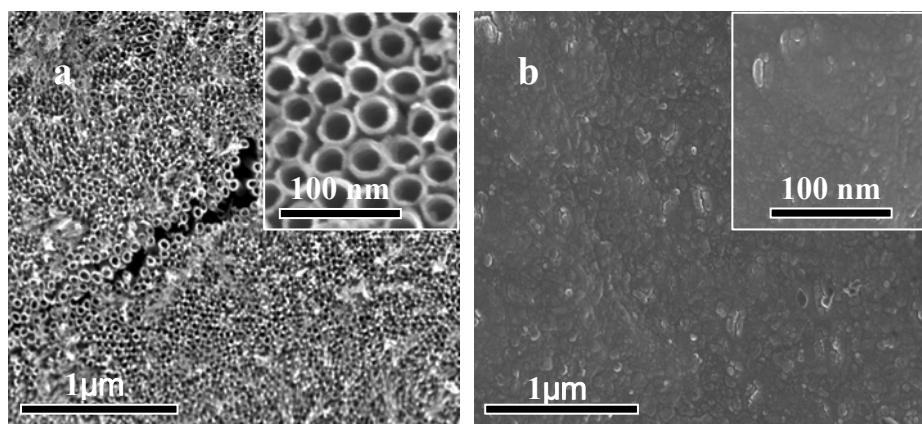


Figure S5. Typical panoramic FE-SEM images of (a) TNTAs and (b) flat anodic TiO₂ layer (FTL) with corresponding enlarged image in the inset.

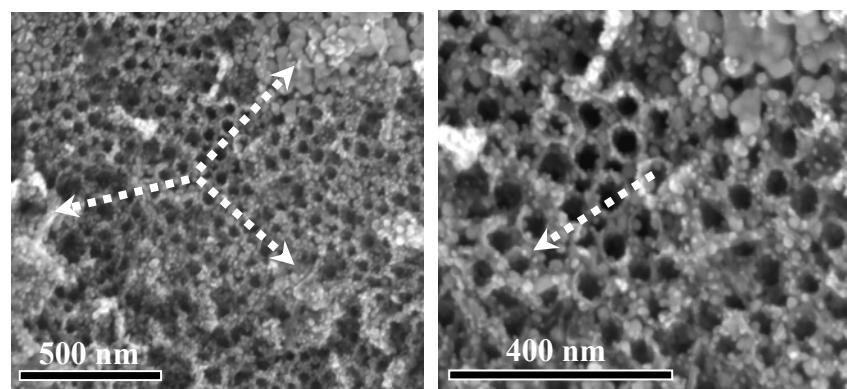


Figure S6. FE-SEM images of GNP/TNTAs (1.14 wt %) after calcination at 450 °C in air for 2 h.

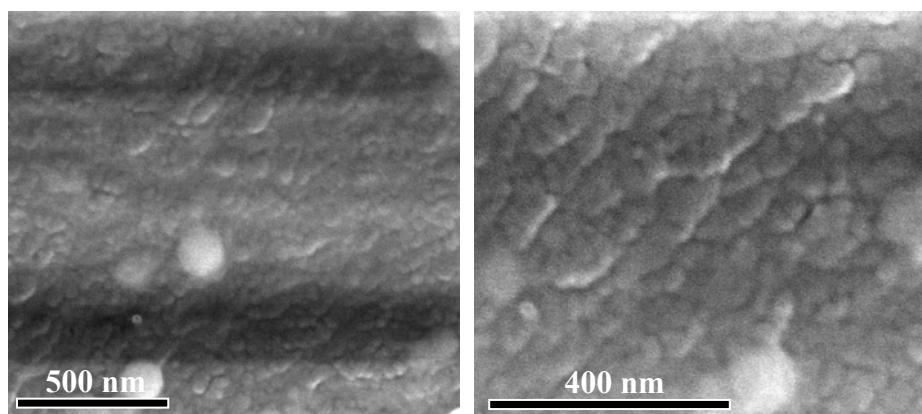


Figure S7. FE-SEM images of GNP/FTL prepared *via* the same self-assembly method.

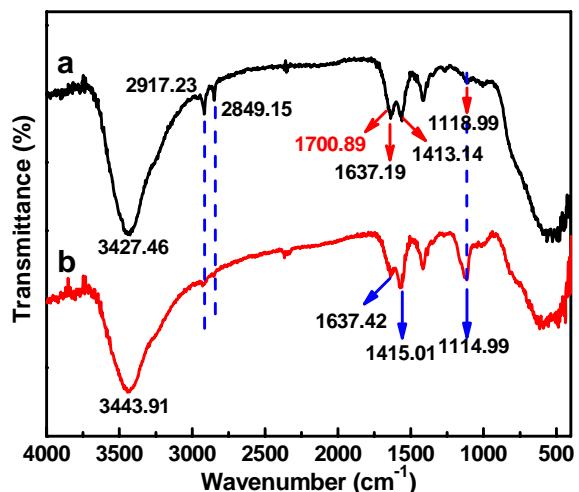


Figure S8. (a) FTIR spectrum of GNP/TNT As (1.14 %). (OH stretching vibration peaks from absorbed H_2O and surface hydroxyl groups of TiO_2 : 3427.46 cm^{-1} , 1637.19 cm^{-1} , 1413.14 cm^{-1} ; C=O stretching vibration peak from MPA: 1700.89 cm^{-1} partly overlapped by 1637.19 cm^{-1} ; Ti-O asymmetric vibration peak: 1118.99 cm^{-1} ; CH_2 vibration peaks from MPA and DDT: 2917.23 cm^{-1} , 2849.15 cm^{-1}), (b) FTIR spectrum of GNP/TNTAs (1.14 %) after calcination at 450 $^{\circ}\text{C}$ in air for 2 h. (OH stretching vibration peaks from absorbed H_2O and surface hydroxyl groups of TiO_2 : 3443.91 cm^{-1} , 1637.42 cm^{-1} , 1415.01 cm^{-1} ; Ti-O asymmetric vibration peak: 1114.99 cm^{-1})

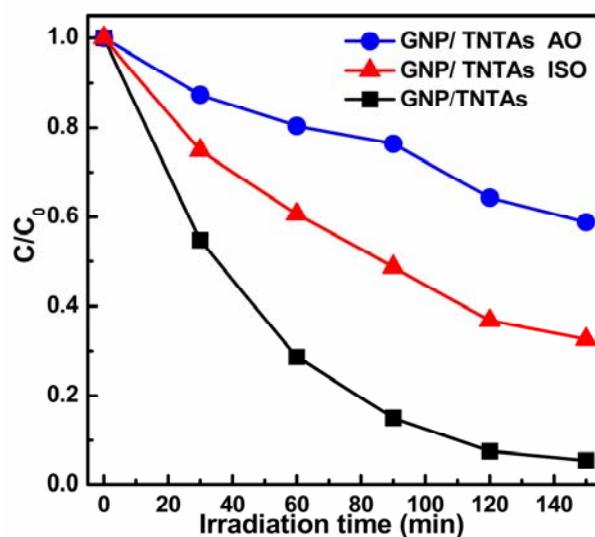


Figure S9. Comparison results of the photocatalytic degradation of MO for GNP/TNTAs (1.14 wt %) with and without the addition of ammonium oxalate (AO, a scavenger for holes) and isopropanol (ISO, a scavenger for hydroxyl radicals).