

## Experimental section

### *Materials*

Hydrochloric acid (AR), methanol (AR) and ethanol (AR) were purchased from China Medicine Group Shanghai Chemical Reagent Company. Imidazole, Rhodamine B (AR), 1, 4-benzoquinone, furfuryl alcohol and dimethyl sulfoxide (99%) were bought from Aladdin Reagent Co. Ltd., Shanghai, China. Carbon nanotubes was supplied by Shandong Dazhan Nanomaterials Co. Ltd..

### *Synthesis of N-doped carbon nanotubes*

The N-doped carbon nanotubes (CNT) was synthesized by thermal treatment of the mixture of HCl purified CNT and imidazole. Specifically, the commercially available CNT was dispersed in concentrated HCl and stirred at room temperature for 12h to remove the exposed metal contaminants. The purified CNT denoted as rCNT was obtained after filtration, washing with deionized water and the subsequent drying. Next, 3.0 g of imidazole was dissolved in 10 mL of ethanol, followed by the addition of 0.5 g of rCNT. Then, the mixture was sonicated for 5 min. Subsequently, ethanol was evaporated at room temperature and the obtained precursor was dried in a 60 °C oven. After ground, the solid mixture was put into a quartz boat and placed in a tube furnace equipped by quartz tube. Afterwards, the mixture was heated to 700 °C with the heating rate of 5 °C/min in the N<sub>2</sub> flow, and maintained for 2 h. The obtained black powder was ground and labeled as NCNT700. NCNT600 and NCNT800 were prepared by the same procedure expect the synthesis temperature set at 600 and 800 °C, respectively.

### *Catalyst Characterization*

The morphology of samples was studied by scanning electron microscope (SEM) and transmission electron microscope (TEM, Tecnai G2 F20) combined with an energy dispersive spectroscopy (EDS) spectrometer. The powder X-ray diffraction (XRD) measurements were carried out on a Rigaku D/Max-2500PC diffractometer with a Cu K $\alpha$  radiation source operating at 50 kV, 300 mA. The specific surface area was

calculated by Brunauer-Emmett-Teller (BET) method, and the nitrogen adsorption-desorption isotherms were acquired from Micromeritics ASAP 2020 HD88 at 77 K. The X-ray photoelectron spectroscopy (XPS) spectra were obtained on an ESCALAB 250 XPS system with a monochromatized Al K $\alpha$  X-ray source (1486.6 eV).

#### *Catalytic performance evaluation*

RhB solution with a concentration of 10 mg/L was prepared by dissolving RhB in ultrapure water, and the degradation of RhB via the activation of PMS in the presence of NCNT700 or other carbon catalysts were carried out in two mode at room temperature. Mode I: 5 mg of NCNT700 and 50 mL of RhB solution were added into a 100 mL bottom rounded flask in sequence and stirred for 60 min. During this stage, RhB was adsorbed on the surface of NCNT700 to reach nearly adsorption equilibrium. Then, 10 mg of PMS was charged to the system to start the reaction. About 2 mL of solution was withdrawn at certain interval and filtrated using a 0.22  $\mu$ m Millipore syringe filter, followed by immediate analysis using a UV-vis spectrophotometer. Mode II: 10 mg of PMS and 50 mL of RhB solution were added into a 100 mL bottom rounded flask in sequence, followed by the quick addition of 5 mg of NCNT700 to initiate the reaction. At certain reaction interval, around 2 mL of solution was withdrawn and filtrated using a 0.22  $\mu$ m Millipore syringe filter, followed by immediate analysis using a UV-vis spectrophotometer. To study the stability of the catalyst, 10 mg of PMS and 50 mL of RhB solution were added into a 100 mL bottom rounded flask in sequence, followed by the addition of 5 mg of NCNT700 to initiate the reaction. When the reaction time reached 20 min, a small amount of the reaction mixture (about 2 mL) was withdrawn and filtrated using a 0.22  $\mu$ m Millipore syringe filter for UV-vis detection. Meanwhile, magnetic stirring was stopped to let the catalyst in the remained mixture gather on the bottom of the flask. Then, the supernatant was removed using a pipette and the recycled catalyst was rinsed with ethanol which was also removed with the assistance of a pipette. Next, the reused catalyst was dried in a 60 oC oven for the second run. In order to collect catalyst for the third run and the subsequent runs, the operation procedure was the same to above approach.

The current response was studied as follows. 10 mg of NCNT700 was dispersed into the mixture containing 1 mL of ethanol and 0.1 mL of Nafion solution (5wt%), which was then sonicated for 1 h to prepare the catalyst ink. Next, 20  $\mu\text{L}$  of the ink was loaded onto a glassy carbon electrode and dried at room temperature. The chronoamperometric test was performed using Ag/AgCl electrodes as reference electrode, and Pt and glassy carbon as the counter and working electrode, respectively. During the measurement, 50 mM  $\text{Na}_2\text{SO}_4$  solution served as the electrolyte and 20  $\mu\text{L}$  of PMS and RhB solution were injected in sequence at certain interval.

Room-temperature electron paramagnetic resonance (EPR, Bruker EMX PLUS) was performed to confirm the role of ROS using 5, 5-dimethyl-1-pyrroline N-oxide (DMPO) and 2, 2, 6, 6-Tetramethyl-4-piperidone hydrochloride (TEMP) as spin trapping agents.

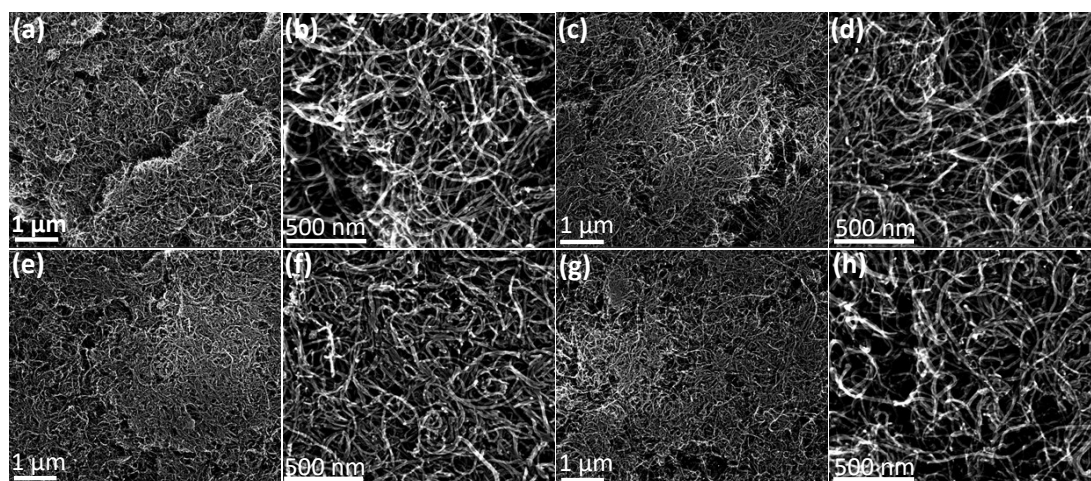


Fig. S1. SEM images of rCNT (a,b), NCNT600 (c,d), NCNT700 (e,f) and NCNT800 (g,h).

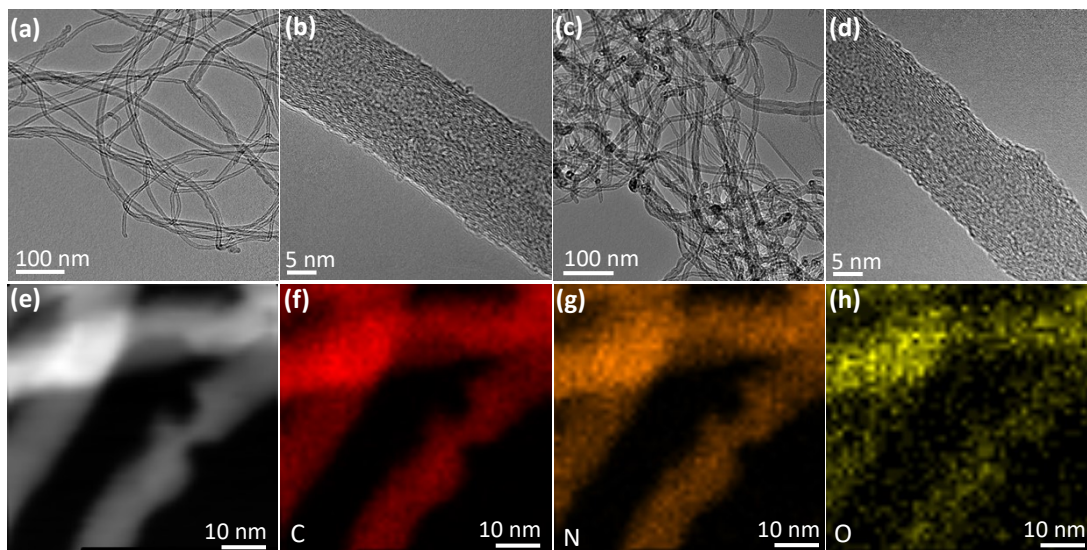


Fig. S2. TEM images of rCNT (a,b) and NCNT700 (c,d). (e-f) was HAADF-STEM and elemental mapping images of NCNT700

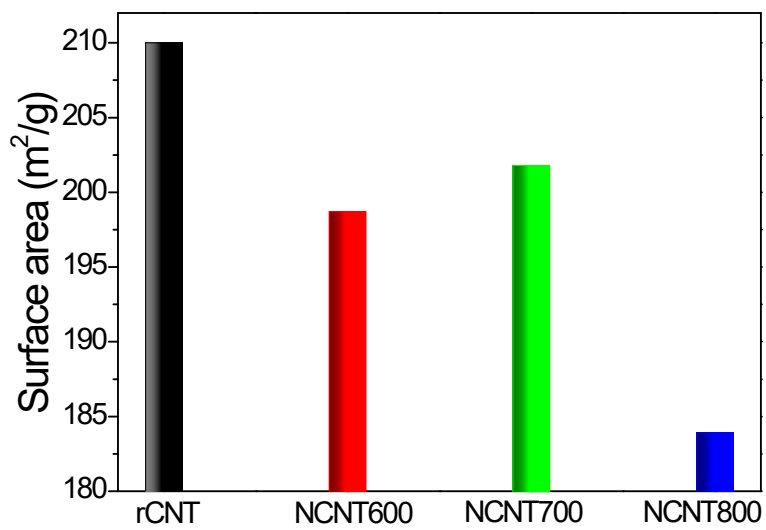


Fig. S3. Specific surface area of each material

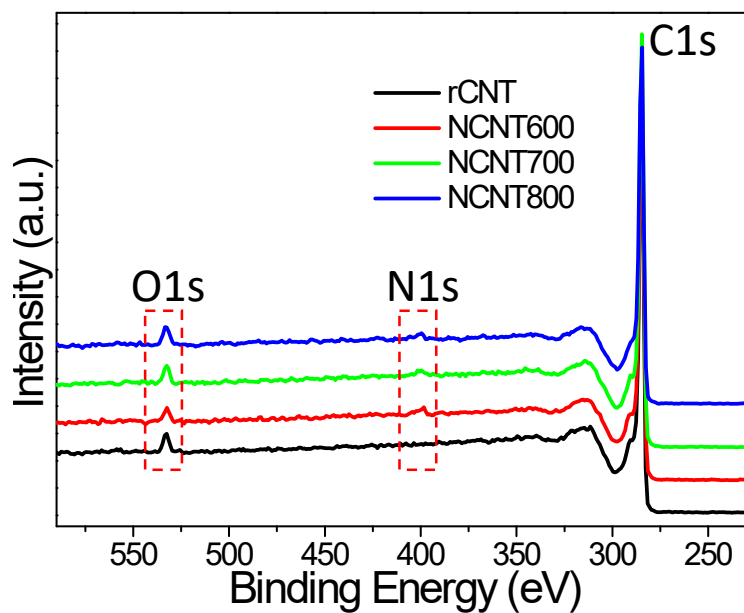


Fig. S4. XPS wide scan spectra of the materials

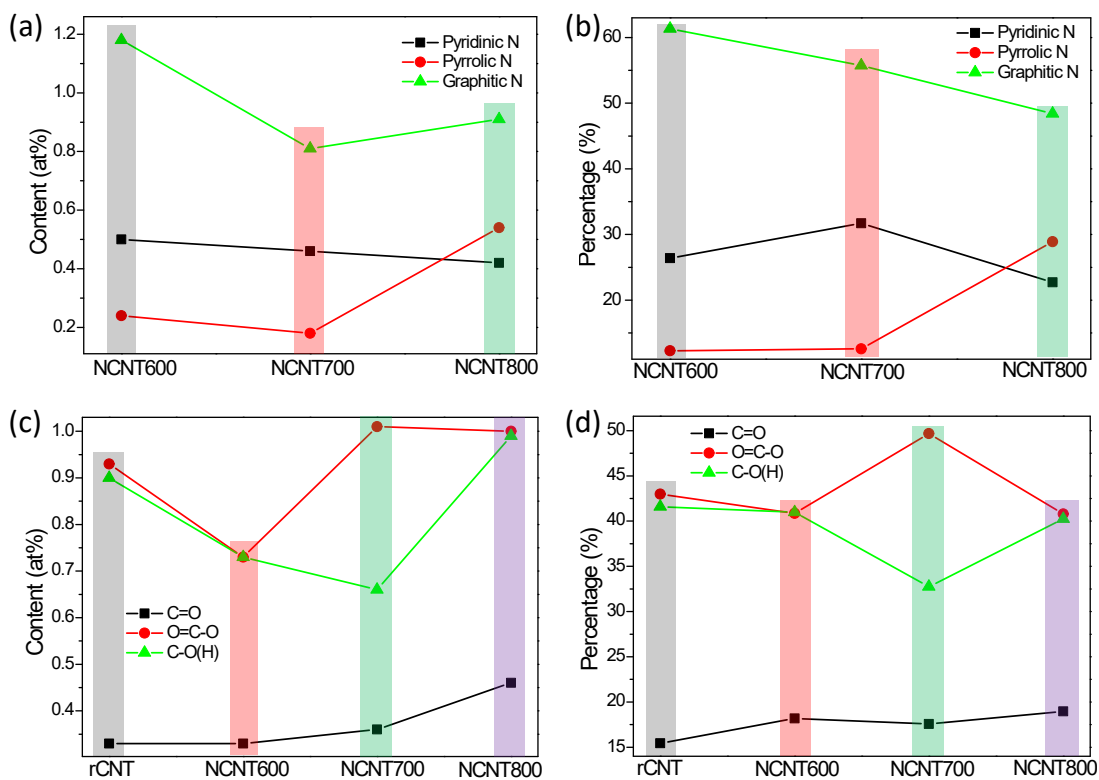


Fig. S5. The content and percentage of each N (a,b) and O (c,d) species on the surface of the materials

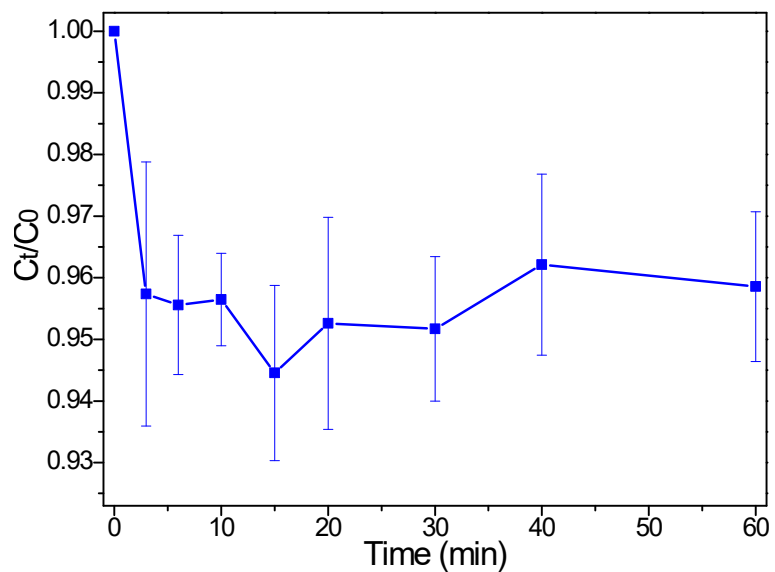


Fig. S6. The adsorption of RhB on NCNT700 in the presence of absolute methanol

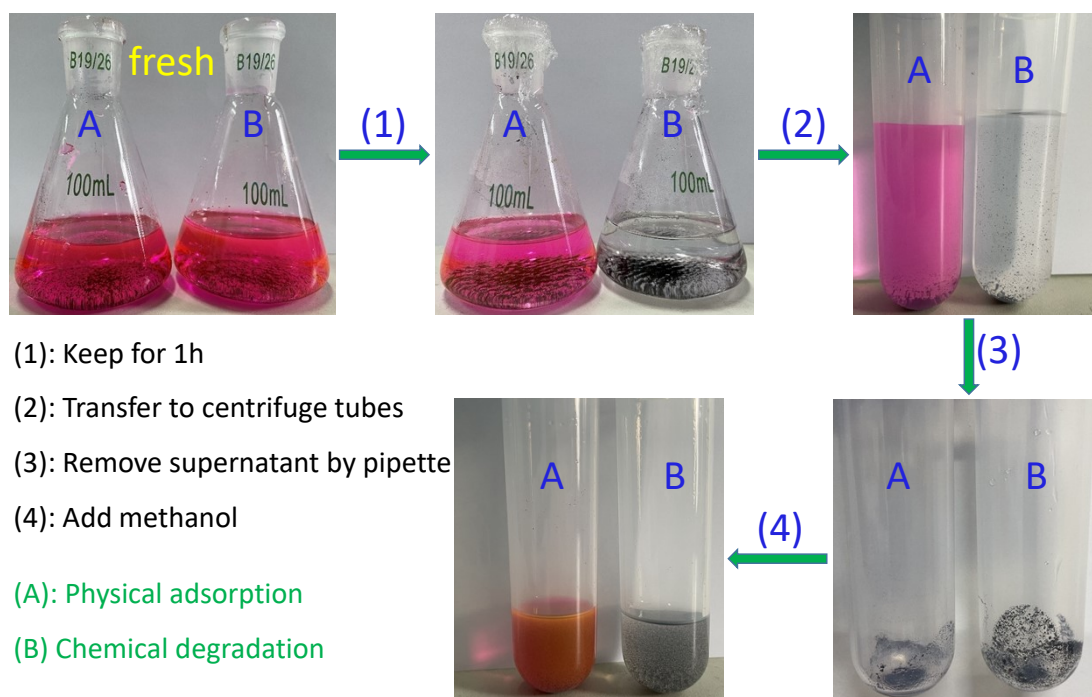


Fig. S7. Comparison of physical adsorption and chemical degradation

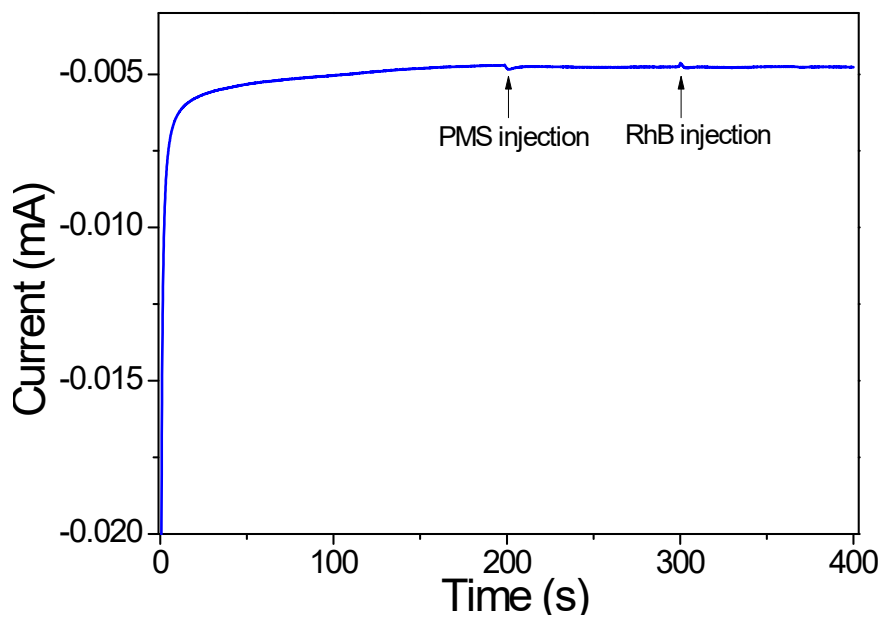


Fig. S8. Chronoamperometric curve upon the addition of PMS and RhB using NCNT700 as working electrode

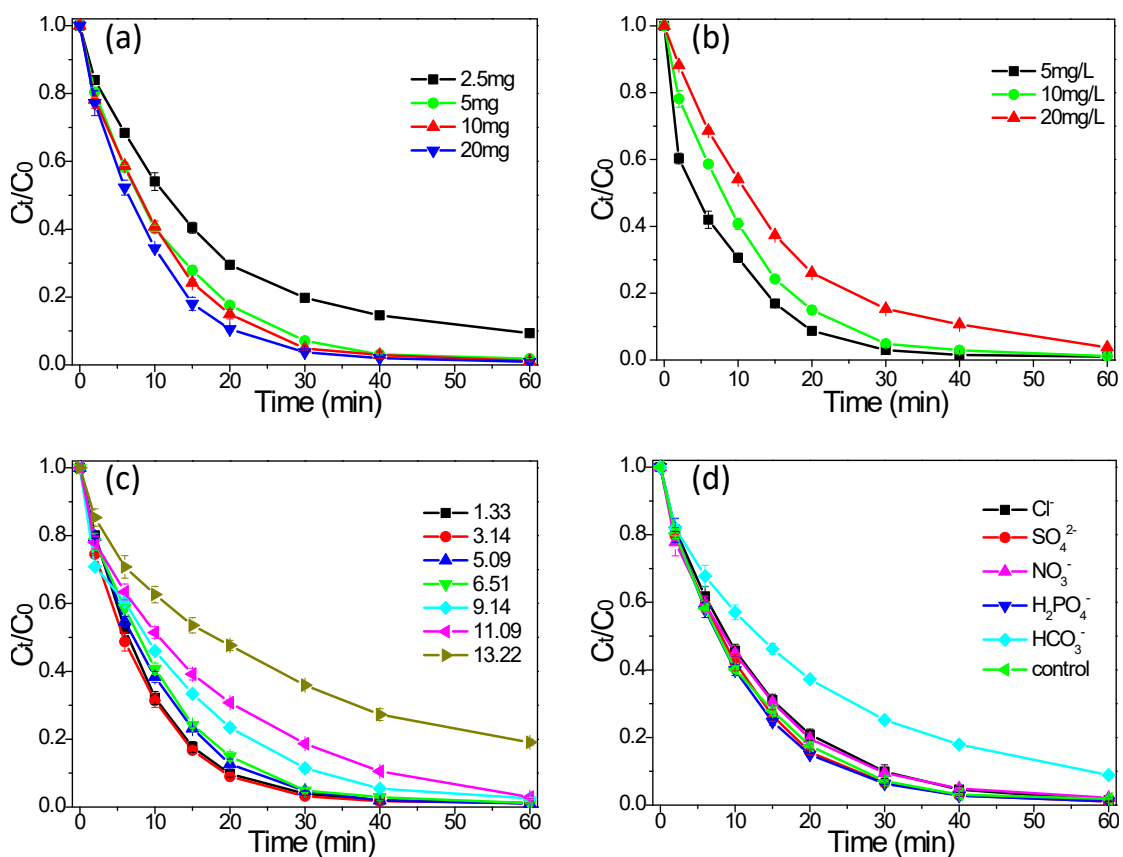


Fig. S9. The effect of PMS dosage (a), RhB concentration (b), initial pH (c) and some typical ions with concentration of 10 mM (d) on the reaction.

Table S1. Surface content of each element detected by XPS and  $I_D/I_G$  ratio

Catalyst	C(at%)	N(at%)	O(at%)	$I_D/I_G$
rCNT	97.84	0	2.16	1.36
NCNT600	96.28	1.92	1.79	1.20
NCNT700	96.52	1.45	2.03	1.13
NCNT800	95.68	1.87	2.45	0.95