**Engineering the Growth of TiO$_2$ Nanotube Arrays on Flexible Carbon Fibre Sheets**

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**Experimental**

Carbon fibre sheet (CFS) was commercially available (purchased from Jiaxing Wanwei Textile Co., Ltd.). All other chemical were purchased from Shanghai Chemical reagents Co. and used as received.

**Growth of TiO$_2$ nanotube arrays on CFS**

Before use, CFS was cleaned with 1 M NaOH solution and distilled water and was dried in an oven at 100 ºC. The clean CFS was immersed into 10 vol.% tetrabutyl titanate/hexane solution for 5 mins, and thus the surface of the CFS was wetted by the solution. When the textile was taken out, hexane would volatilize rapidly and only tetrabutyl titanate would be left on the texture. Then, the textile was exposed to water vapor to promote the hydrolysis of tetrabutyl titanate and the generation of TiO$_2$. This wetting-hydrolysis process was repeated several times in order to increase the loading capacity of TiO$_2$. After that, the textile coated by TiO$_2$ was rolled up and placed into an autoclave filled with 10 M NaOH solution. The size of the textile is dependent on the volume of the autoclave. For a 50 mL autoclave, the texture can be several tens of centimeters in length. For a 1 L autoclave, the textile can be several meters in length. After heated at 160 ºC for 48 hours, the autoclave was cooled down to room temperature and the textile was taken out, washed alternatively with 0.1 M HAc and distilled water five times, and calcined at 400 ºC under air atmosphere for 5 hours. It could be seen that the CFS had changed its color from the initial black to white, which suggested the presence of TiO$_2$.

**Characterizations of TiO$_2$ nanotube arrays on the CFS**

The loading capacity of TiO$_2$ on the textile was determined by comparison of the initial weight and the final weight of the textile. Scanning electron microscopy (SEM) images were taken with a Hitachi S-4700 field emitting scanning electron microscope. Transmission electron microscope (TEM) image, high resolution TEM (HRTEM) image, and selected area electron
diffraction (SAED) pattern were taken with a FEI Tecnai Tecnai G2 F20 electron microscope. X-ray diffraction patterns were recorded on an X’Pert PRO SUPER rA rotation anode X-ray diffractometer with Ni-filtered Cu Kα radiation (λ=1.54178 Å). Diffuse reflection spectra were measured by a Shimadzu 3150 UV-vis-near-infrared spectrophotometer equipped with an integrating sphere. The N₂ adsorption-desorption isotherms and BET surface area were measured on a Micromeritics ASAP 2020 instrument. The samples for SEM, XRD, DRS, and gas adsorption characterizations were the textiles directly. The samples for TEM and HRTEM characterizations were prepared as follows: TiO₂ nanotube (TiNT) arrays were exfoliated from the texture by ultrasound, dispersed in absolute ethanol, and dropped onto a carbon film supported on a copper grid.

**Adsorbability of CFS grown by TiNT arrays towards dyes dissolved in water**

Rhodamine B (RhB) solution (concentration: 1.0 mg/L) and fluorescein solution (concentration: 2.0 mg/L) were used to assess the adsorbability of CFS grown by TiNT arrays. Each dye solution (volume: 30 mL) was immersed by a piece of the textile with or without the growth of TiNT arrays (dimensions: 2 cm × 2 cm). Next, the solution was kept in a dark environment for 12 hours. The textiles with TiNT arrays faded the dye solutions obviously while those without TiNT arrays did not. The concentration changes of the dye solutions were monitored by UV-vis absorption spectroscopy (UV3150, Shimadzu). RhB has a maximum absorption around 550 nm.

**Evaluation of photocatalytic activity of CFS grown by TiNT arrays**

RhB is a chemically stable and poorly biodegradable dye contaminant in wastewater. Here we use its decomposition as a simulation to demonstrate the great advantages of CFS modified by TiNT arrays in the purification of contaminated water. All the experiments were carried out at room temperature (~15 °C). Unless otherwise indicated, CFS modified by TiNT arrays for photocatalytic experiments are synthesized via three times of wetting-hydrolysis cycles.

At first, we investigated the robustness and reusability of CFS modified by TiNT arrays by repeated adsorption-lifting-irradiation experiments. A piece of the textile (dimensions: 2 cm × 2 cm) was immersed into 50 mL RhB solution (concentration: 7.5 mg/L) for 30 minutes. The textile was lifted from the solution and irradiated by a 10 W UV lamp (wavelength: 254 nm). During the irradiation, the textile was placed 10 cm away from the lamp, and its each side was
irradiated 15 minutes. RhB molecules on the surface of the textile were monitored by measuring the diffuse reflection spectrum. The above adsorption-lifting-irradiation process was repeated twenty times.

Subsequently, we investigated the photodegradation behaviors of RhB under the continuous photocatalysis of the textile. During these studies, the textile was immersed in the solution all the time. The concentration of RB solution was monitored by detecting the absorbance at 550 nm per hour. For a comparison, the photocatalytic activities of equal sizes of pure CFS and the modified CFS synthesized via seven times of wetting-hydrolysis cycles were also investigated.

**Scheme S1.** Schematic description of the strategy of the engineering growth of TiNT arrays on CFS.

**Fig. S1** (a) Optical photograph of CFS. (b) SEM image of CFS before the coating of TiO₂. The surface of the textile is clean. (c, d) SEM images of CFS coated by TiO₂ through the wetting-hydrolysis process. The surfaces of the textiles are covered by TiO₂ particles. There are a small amount of TiO₂ particles on the surface of the textile when the wetting-hydrolysis process is not repeated (1c). A large amount of TiO₂ particles are seen when the wetting-hydrolysis process is repeated three times (1d).
**Fig. S2** XRD patterns. (a): pure CFS; (b) CFS grown by TiNT arrays.

**Fig. S3** SEM images show that the covering degree of TiNT arrays can be adjusted by repeating the wetting-hydrolysis process. (a, b): no repetition; (c, d): seven times of repetition. When the wetting-hydrolysis process is not repeated, only sparse and short nanotubes are seen. When the process is repeated seven times, every fibre in the CFS is overlaid with dense tubes. More interestingly, the nanotubes seem to self-assemble into ribbons.

In Fig. S3a, the growth of TiNT arrays does not cause a detected change of the weight of the textile. But in Fig. S3c, one square centimeter of CFS supports 8 mg of TiNT arrays.
**Fig. S4** UV-vis absorption spectra of the dye solutions before and after interaction with CFS grown by TiNT arrays, where insets are the optical photographs of the dye solutions. (a): RhB solution; (b): fluorescein solution. Benefiting from the mesoporosity and the large surface area, the CFS grown by TiNT arrays exhibits a strong adsorbability towards the dyes, as testified by the fading phenomenon of the dye solutions in a dark environment. Under the same conditions, equal sizes of pure CFS does not fade the dye solutions obviously.

**Fig. S5** Diffuse reflection spectra of CFS modified by TiNT arrays after twenty times of adsorption (red line) and irradiation with UV light (blue line). It can be seen that the textile still has a strong adsorobility towards RhB and a high photocatalytic activity.
**Fig. S6** Representative evolution of UV-vis absorption spectra of RhB solution under the continuous photocatalysis of the modified CFS synthesized via seven times of wetting-hydrolysis cycles (a) and three times of wetting-hydrolysis cycles (b). (c) Corresponding photodegradation curves.

Obviously, the textile carrying more TiNT has a higher photocatalytic activity. By the way, pure CFS has no photocatalytic activity; it only causes a slight fading of RhB solution due to the adsorption of a small amount of RhB by the textile.