Supporting Information

1. Synthesis of protonated titanate nanotube:

The H-TDNT nanotubes were prepared using the alkaline hydrothermal synthesis. Briefly, 1.0 g of TiO_2 powder was dispersed into 30 mL of 10 M NaOH aqueous solution under stirring. After stirring for 24 h, the alkaline suspension was transferred into an autoclave with Teflon inner line and statically heated at 150 °C for 48h. The H-TDNT nanotubes were recovered by washing with 0.1 M dilute HCl solution and deionized water till neutral.

2. Photocatalytic reaction test

The photocatalytic activity of N-TDNT-450 was evaluated by measuring the decomposition of methylene blue under visible light irradiation. A 300 W xenon lamp (Trusttech, Beijing, China) with Uv-cutoff filter was used as the visible light source. For a photocatalytic experiment, 100 mg catalyst was dispersed into 100 mL MB aqueous solution (10 ppm) in a Pyrex glass beaker. The distance between the beaker and the light source is about 20 cm. The suspensions containing MB and catalysts were magnetically stirred for 60 min in the dark prior to irradiation, for establishing absorption–desorption equilibrium. The absorbance of MB solution was determined using Cintra 10 **UV**-visible spectrometer. The concentration of MB was obtained according to the linear relation between the absorbance (at 661.0 nm) and the concentration of MB solution.

3. Possible formation mechanism of N-TDNTs using ammonia salts as N-sources:

After annealing over 350 °C, N-doped anatase nanotubes can be produced despite of the N-sources used in the solvothermal process except for NH_4Cl , suggesting the ammonia salts were thermal decomposed to produce ammonia under the solvothermal conditions. The in-situ produced ammonia or NH_4^+ will react with H-TDNT to form NH_3 -TDNTs or NH_4 -TDNT that transfer to N-doped anatase in annealing.

4. Fig S1. XRD-Patterns of N-TDNT synthesized through solvothermal route with different nitrogen sources

The XRD results indicate that only ammonia chloride in these cases can promote the anatase phase formation in the solvothermal treatments, other ammonia salts used here prefer to form ammonia titanates or mixture of H-TNT and nanotubular ammonia titanates.



Fig. S1. XRD patterns of N-TDNT synthesized with different nitrogen sources