Supplementary Information for

(NH$_4$)$_2$SO$_4$-assisted polycondensation of dicyandiamide for porous g-C$_3$N$_4$ with enhanced photocatalytic NO removal

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- Experimental Section

1.1 Preparation of bulk g-C$_3$N$_4$ and (NH$_4$)$_2$SO$_4$-modified g-C$_3$N$_4$

All chemicals were reagent grade and used without further treatment. The bulk g-C$_3$N$_4$ (CN) was prepared by the polycondensation of dicyandiamide (2g) loaded in a crucible at 550 °C for 2 h in a muffle furnace. The ramping rate was 5 °C /min. The sample was cooled naturally to room temperature. To prepare (NH$_4$)$_2$SO$_4$-modified g-C$_3$N$_4$, 2g of dicyandiamide powder and different amount of (NH$_4$)$_2$SO$_4$ were dissolved with 30 mL water in an alumina crucible. The solution of dicyandiamide and ammonium sulfate were then dried at 60 °C overnight to get the composite precursors. The composite precursors loaded in a crucible were heated to 550 °C at a heating rate of 5 °C min^{-1} in a mu□e furnace and maintained for 2 h. According to the molar ratios of dicyandiamide: (NH$_4$)$_2$SO$_4$ (2:1, 2:1.5, 2:2, and 2:2.5), these samples were named as CN-1, CN-1.5, CN-2 and CN-2.5, respectively.

1.2 Characterization

The X-ray diffraction (XRD) patterns of the samples were recorded on an X-ray diffractometer equipped with intense Cu Kα radiation (Model D/max RA, Rigaku Co., Japan). The morphological structure was analyzed using scanning electron microscope (SEM, JEOL model JSM-6490, Japan) and transmission electron

microscope (TEM, JEM-2010, Japan). The Brunauer–Emmett–Teller (BET) specific surface area (SBET) of the samples were determined using a nitrogen adsorption apparatus (ASAP 2020, USA) with all samples degassed at 100 °C for 12 h prior to measurements. X-ray photoelectron spectroscopy (XPS) measurement was carried out to investigate the surface chemical compositions and states with Al Kα X-ray (hν = 1486.6 eV) radiation source operated at 150 W (Thermo ESCALAB 250, USA). The UV-vis diffuse reflection spectra (UV-vis DRS) were obtained for the dry-pressed disk samples by using a Scan UV-vis spectrophotometer (UV-2450, Shimadzu, Japan) with 100% $BaSO_4$ as the standard sample. The photoluminescence spectra (PL) for the samples were measured with a fluorescence spectrophotometer (FS-2500, Japan) using a Xe lamp as excitation source with optical filters.

1.3 Visible light photocatalytic capability for NO purification

The visible light responsive photocatalytic activity was evaluated by removal of NO at ppb-level in a continuous flow reactor. The reactor had a capacity of 4.5 L (30×15×10 cm), made of polymeric glass and covered with Quartz-Glass. A commercial tungsten halogen lamp (150 W) was vertically placed outside and 20 cm above the reactor. A UV cut-off filter (420 nm) was used to remove UV light from the lamp. The as-prepared sample (0.20 g) was dispersed in distilled water (50 mL) in two beakers via ultrasonic treatment for 10 min and then spread on two glass dishes. The coated dishes were dried at 60 °C and were placed at the center of the reactor after being cooled to room temperature. The NO gas was acquired from a compressed gas cylinder at a concentration of 100 ppm of NO (N_2 balance). The initial concentration of NO was diluted to about 600 ppb by air streaming. The flow rates of the air stream and NO were controlled at 2.4 L min^{-1} and 15 mL min^{-1}, respectively. The gas streams were premixed completely by a gas blender. The relative humidity was controlled at 50% in the air stream. When the adsorption−desorption equilibrium was achieved, the lamp was turned on. The concentration of NO was measured every minute by a NOx analyzer (Thermo Scientific, 42i-TL), which also monitored the concentrations of NO, NO_2 and NOx. The removal ratio (η) of NO was calculated using η(%)=(1−C/C_0)×100%, where C and C_0 represents the outlet and inlet concentration of NO.