Supplementary Information for

Scalable and Clean Exfoliation of Graphitic Carbon Nitride in NaClO Solution: Enriched Surface Active Sites for Enhanced Photocatalytic H_2 Evolution

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Part 1 The detailed experimental information of all ESR, EIS and

Photocurrents test

a. ESR signals were investigated on a Bruker model A300 electron paramagnetic resonance spectrometer at room temperature with a 300W Xe lamp as the visible light source: microwave frequency = 9.852 GHz, microwave power = 20.0 mW, modulation amplitude = 4.00G, and sweep time = 60 s. Specifically, bulk powders were packed into an ESR tube, and then under light exposure, the accumulation of conditions was three continuous scans to superposition (g = 2.0035; trace oxygen in ESR tube originated from the air).

b. Electrochemical impedance spectroscopy (EIS) was recorded by a Chi660e electrochemical workstation based on a conventional three-electrode system from the frequency 0.01 Hz to 100 kHz at the open circuit potential. Each time, 0.009 g of sample powder and 0.01 g of polyvinylidene fluoride (PVDF) were uniformly ground and mixed with a mortar, and then added with about 20 drops of N-methylpyrrolidone. The mixture was magnetically stirred for 5 h, then evenly spread on a 1 × 1 cm^2 indium tin oxide (ITO) glass, and dried for further use. A standard three-electrode system adopts platinum wire, ITO glass and Ag/AgCl (saturated KCl solution) as the counter electrode, working electrode and reference electrode, respectively. All the EIS measurements were performed at a constant potential of 0.0 V and with 0.5 M Na_2SO_4 solution as the supporting electrolyte. In the experiment using electrochemical workstation, each sample measurement showed different open-circuit voltages in the conventional three-electrode system. For the experimental stability and based on relevant references and related electrode parameters, we set all the electrochemical workstation measurements at a constant potential of 0.0 V.

c. Photocurrents were measured with an electrochemical analyzer (Chi660e, Chen Hua Instruments, Shanghai, China). A standard three-electrode system adopts platinum wire, indium tin oxide (ITO) glass and Ag/AgCl (saturated KCl solution) as the counter electrode, working electrode and reference electrode, respectively. In each experiment, 0.009 g of sample powder and 0.01 g of polyvinylidene fluoride (PVDF) were uniformly ground and mixed with a mortar, and then added with about 20 drops

of N-methylpyrrolidone. The mixture was magnetically stirred for 5 h, then evenly spread on a 1 × 1 cm^2 ITO glass, and dried for further use. All the photocurrent measurements were performed under the illumination of a 300 W Xe lamp (λ > 420 nm), with 0.5 M Na_2SO_4 solution as the supporting electrolyte and at the applied voltage 0.0 V.

Part 2 AFM images of the exfoliated NaClO g-C_3N_4 nanosheets

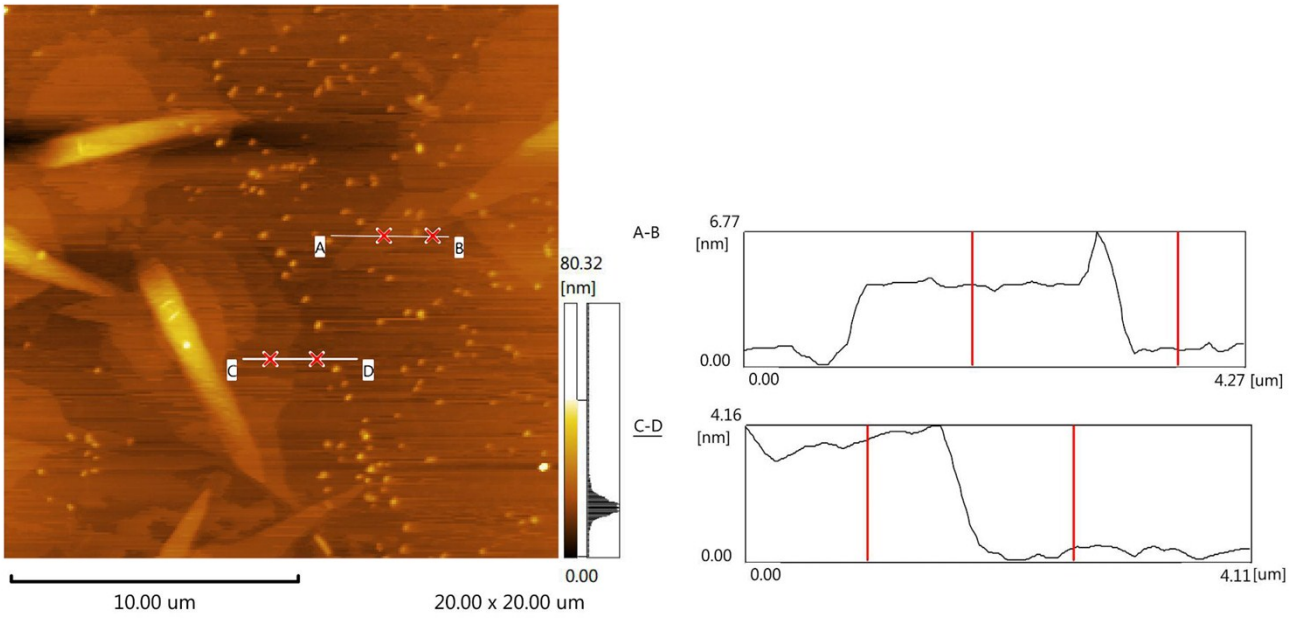

Figure S1. AFM image and corresponding height profile of the exfoliated NaClO g-C_3N_4 nanosheets.

Atomic Force Microscope (AFM) images were acquired in tapping mode on Japan Shimadzu SPM9700. AFM results (Figure S1) clearly present sheet-like objects of ~ 10 um in lateral size. Despite some overlaps observed in the images, the ultrathin few-layer is predominant and the measured average thickness is ~ 4 nm, which indicates the measured sheets are few-layer nanosheets, as the theoretical thickness of g-C_3N_4 is 0.33 nm [*Adv. Mater.* 2014, 26, 4438–4443.]. This finding directly proves g-C_3N_4 has been effectively delaminated into its few-layer sheets. At present, the AFM results have been added in the Supplementary Information as Figure S1, you can see the related discussions on page 4 of the revised manuscript.

Part 3 XRD and UV-vis spectrum of the reused NaClO g-C_3N_4 sample

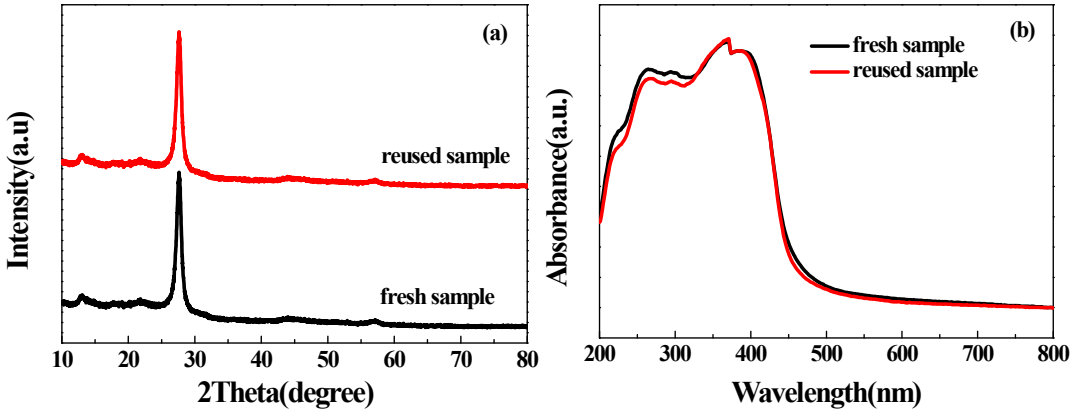

Figure S2. XRD patterns (a) and UV–vis absorption spectra (b) of the reused NaClO g-C_3N_4 under visible light irradiation.