Supporting Information

Superior photocatalytic hydrogen peroxide production over g-C_3N_4 with $Ni(OH)_2$ as an oxidation

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1 Experiment

1.1 Reagents:

Melamine, urea, potassium hydrogen phthalate ($C_8H_5KO_4$), nickel acetate ($Ni(CH_3COO)_2 \cdot 4H_2O$), sodium carbonate ($Na_2CO_3$), silver nitrate ($AgNO_3$), isopropanol (IPA), p-benzoquinone (BQ), and anhydrous ethanol (C_2H_5OH) were purchased from Sinopharm. Potassium iodide (KI) was purchased from Macklin. All chemical reagents are of analytical purity and used directly without further purification.

2.2 Characterization.

The phase composition of the as-prepared catalysts was analyzed via an X-ray diffractometer (XRD, Empyrean, Netherlands) with a 2θ range of 10–80° and a scanning rate of 5°/min. The surface morphology of the samples was observed using a field emission scanning electron microscope (FESEM, ZEISS GeminiSEM 300, Germany) and a field emission transmission electron microscope (FETEM, FEI Tecnai G2 F30). The chemical composition and surface electronic states of the catalysts were investigated by X-ray photoelectron spectroscopy (XPS, Thermo Scientific K-Alpha, USA). N_2 adsorption-desorption isotherms were measured on a Brunauer-Emmett-Teller (BET) analyzer (Micromeritics ASAP 2460) to evaluate the specific surface area and pore structure. The optical absorption properties of the photocatalysts were recorded on a UV-visible (UV-VIS) spectrophotometer (Shimadzu UV-2600) in the wavelength range of 200–800 nm. The charge transfer and separation efficiency were assessed by a steady-state/transient photoluminescence (PL) spectrometer (FLS1000,

Edinburgh Instruments, UK).

2.3 Free radical and hole trapping experiments.

To elucidate the mechanism of photocatalytic H_2O_2 production, active species trapping experiments were performed. Ethanol, $AgNO_3$, benzoquinone (BQ), and isopropanol (IPA) were employed as quenchants to capture holes (h^+), electrons, superoxide anion radicals ($\cdot O_2^-$), and hydroxyl radicals ($\cdot OH$), respectively. Furthermore, $\cdot O_2^-$ radicals were directly analyzed by electron paramagnetic resonance spectroscopy (EPR, Bruker ESR A300, Germany) at room temperature (298 K), using 5,5-dimethyl-1-pyrroline N-oxide (DMPO) as the spin-trapping agent.

2.4 Photoelectrochemical test.

The photoelectrochemical properties of the obtained samples were investigated using an electrochemical workstation (PGSTAT204, Metrohm Autolab) equipped with a standard three-electrode system, under the conditions of an open-circuit voltage of 0.6 V and a frequency range of 0.01–100,000 Hz. In this system, FTO glass served as the working electrode, 0.1 M Na_2SO_4 aqueous solution as the electrolyte, a platinum wire electrode (approximately 1 cm × 1 cm) as the counter electrode, and a calomel electrode as the reference electrode (note: platinum electrode is typically used as counter electrode, corrected for rationality). All measurements were carried out under 420 nm visible-light irradiation, including electrochemical impedance spectroscopy (EIS), transient photocurrent response, and Mott-Schottky plots. The preparation procedure of the working electrode was as follows: 20 mg of the photocatalyst was

dispersed into a mixed solution consisting of 480 μL ethanol and 20 μL Nafion, followed by ultrasonication for 30 minutes to form a homogeneous suspension. Subsequently, 100 μL of the suspension was uniformly coated onto a clean FTO glass substrate (effective area: 20 × 20 mm) and dried at 60 °C to obtain the working electrode.

2.5 Apparent quantum yield (AQY) measurement

The apparent quantum yield (AQY) for the catalysts was tested under the same reaction conditions (using a 420 nm LED lamp as light source). AQY was calculated by the following Eq. (1) and (2):

The number of incident photons (mol): $N = \frac{E\lambda}{h\nu}$ (1)

$$AQY = \frac{2 \times H_2O_2\ formed\ (mol)}{N\ (mol)} \times 100\% \quad (2)$$

2 Results

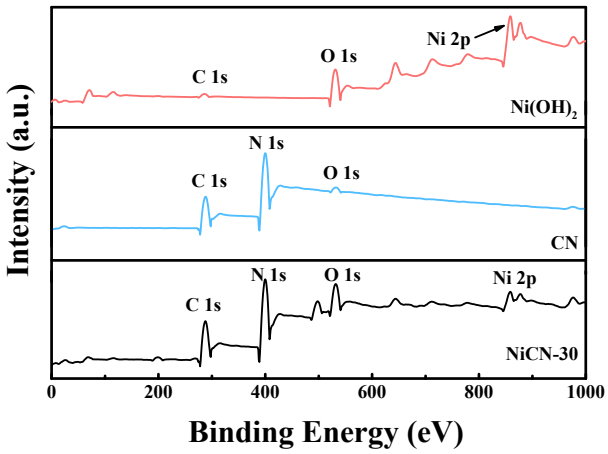

Fig. S1 XPS survey spectra for $Ni(OH)_2$, CN, and NiCN-30

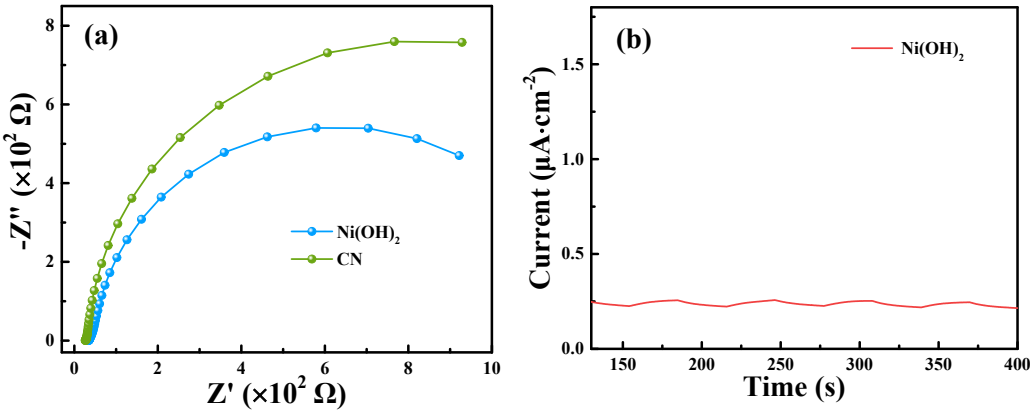

Fig. S2 EIS Nyquist plots (d) of CN and $Ni(OH)_2$ (a), photocurrent-time curve of $Ni(OH)_2$ (b).

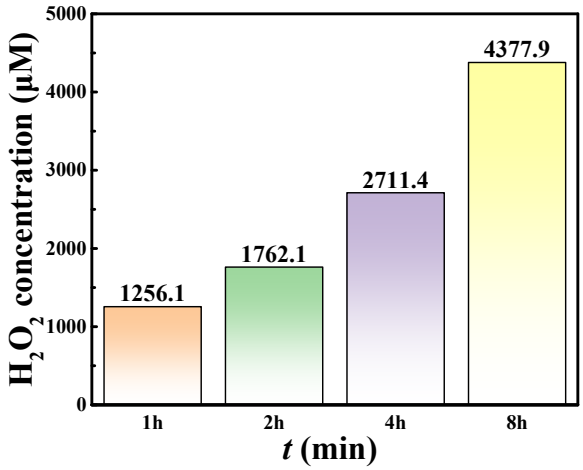

Fig. S3 The cumulative H_2O_2 yield on NiCN-30 during 8 hours of continuous irradiation

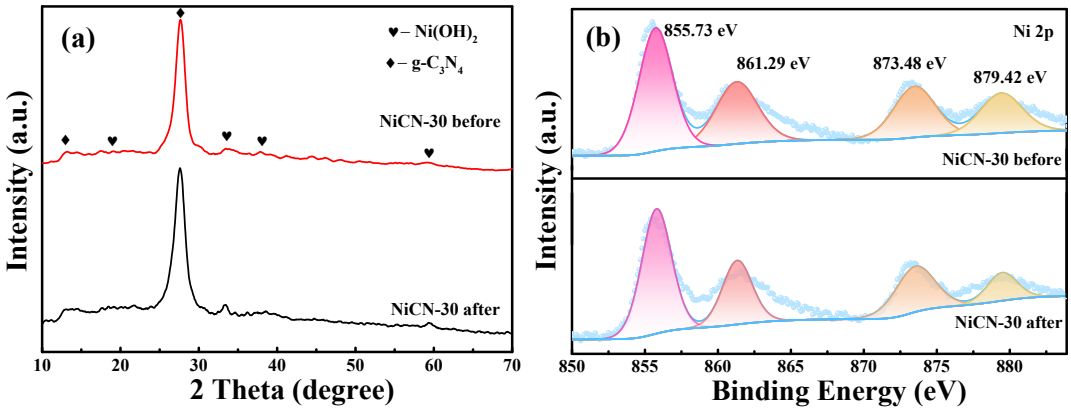

Fig. S4 XRD patterns (a) and high-resolution XPS spectra of Ni 2p for NiCN-30 before and after reaction