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Optimizing electronic configuration of h-BN for boosting photocatalytic transformation of acid gases under visible light

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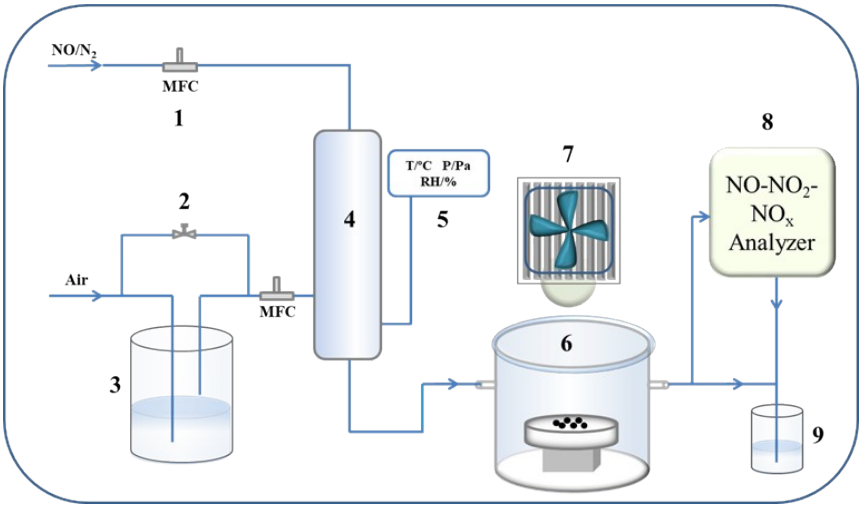
1. Material characterization

Powder X-ray diffraction (XRD) patterns were obtained on a Rigaku diffractometer (MiniFlex 600) with Cu Kα at 40 kV. Fourier transform infrared spectroscopy (FTIR) spectra were recorded on a Thermo Nicolet iS50 Spectrometer with DTGS ATR detector. X-ray photoelectron spectroscopy (XPS, Thermo Scientific ESCALAB 250XI) was used to identify the surface chemical composition of samples. The scanning electron microscope (SEM) measurements were performed on a Hitachi S-8010 field-emission scanning electron microscope with an accelerating voltage of 5 kV. The transmission electron microscope (TEM) measurements were performed on a FEI talos F200s electron microscope equipped with an energy dispersive X-ray detector (EDX) at an acceleration voltage of 100 kV. The N_2 adsorption-desorption isotherms plots of samples were obtained by a Micromeritics ASAP 2020 analyzer and the specific surface area was calculated by Brunauer-Emmet-Teller (BET) method. The electron paramagnetic resonance (EPR) spectroscopy was conducted by a BRUKER A300 spectrometer. The light absorption performances were investigated by a UV-vis spectrometer (SolidSpec-3700, Shimadzu, Japan). The photoelectrochemical testing was performed by Bio-Logic VSP-300 electrochemical workstation. The temperature-programmed desorption curves of gas were recorded by a micromeritics AutoChem II 2920 Chemisorption analyzer.

2. Photocatalytic NO removal process

The photocatalytic NO removal process is shown in **Scheme S1**. The surface dish containing 20 mg of catalyst was placed in a closed quartz reactor and an LED

lamp was used as the light source. Using standard air to dilute NO to 1 ppm and control the flow rate of mixed gas at 500 $mL \cdot min^{-1}$. The standard air was first bubbled through a bottle containing water before being mixed with NO. Before the illumination, the feed gas was passed in the dark state for a while until the NO concentration was stable ensuring that the catalyst surface reached the equilibrium of adsorption and desorption. A NO-NO_2-NO_x Analyzer (U. S. A. EPA, Model 42i) was used for product analysis. The removal efficiency (η) of NO was calculated as $\eta = (1-C/C_0) \times 100\%$, where C and C_0 are the transient and initial concentrations of NO, respectively.



Scheme S1 Schematic diagram of the photocatalytic NO removal process.

1 - mass flow controller; 2 - ball valve; 3 - bubbler; 4 - gas mixing tank; 5 - temperature and humidity detector; 6 - reactor; 7 - light source; 8 - NO-NO_2-NO_x analyzer; 9 - tail gas collector.

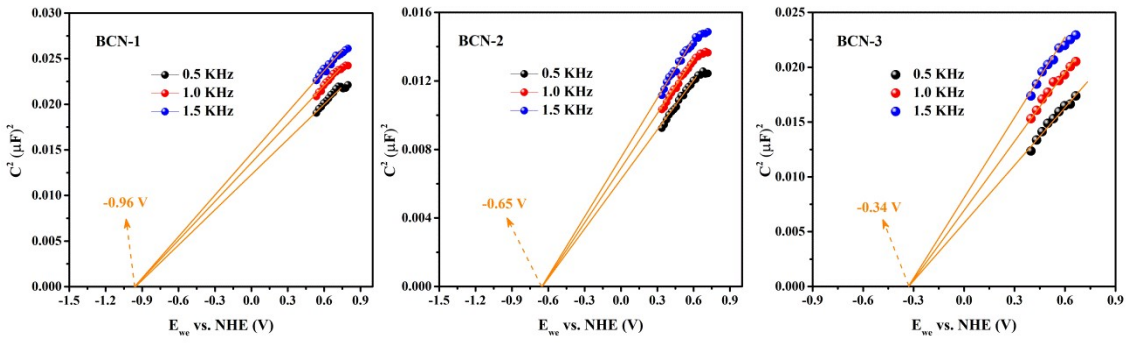


Fig. S1 Mott-Schottky plots of BCN samples.

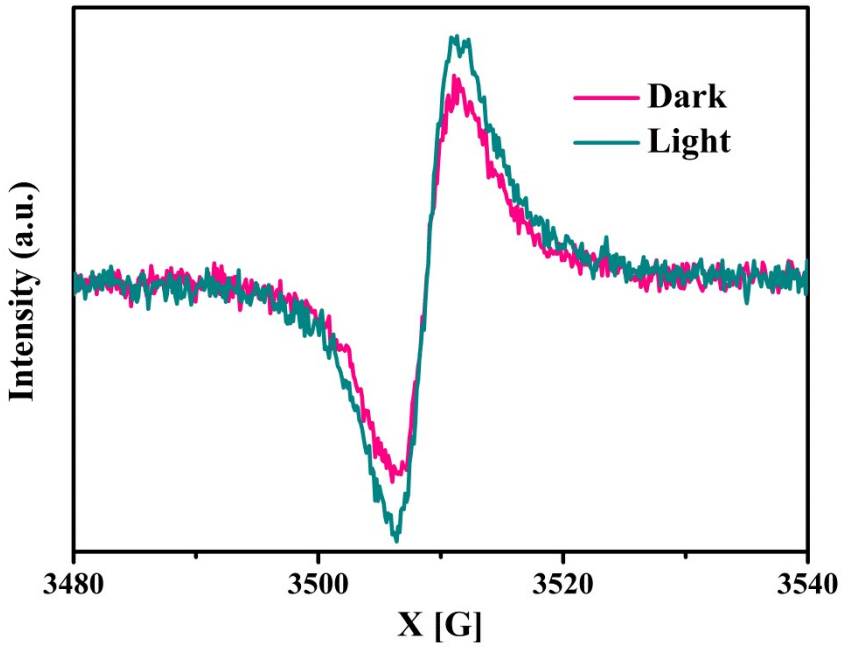


Fig. S2 EPR pattern of BCN-2 under visible illumination.

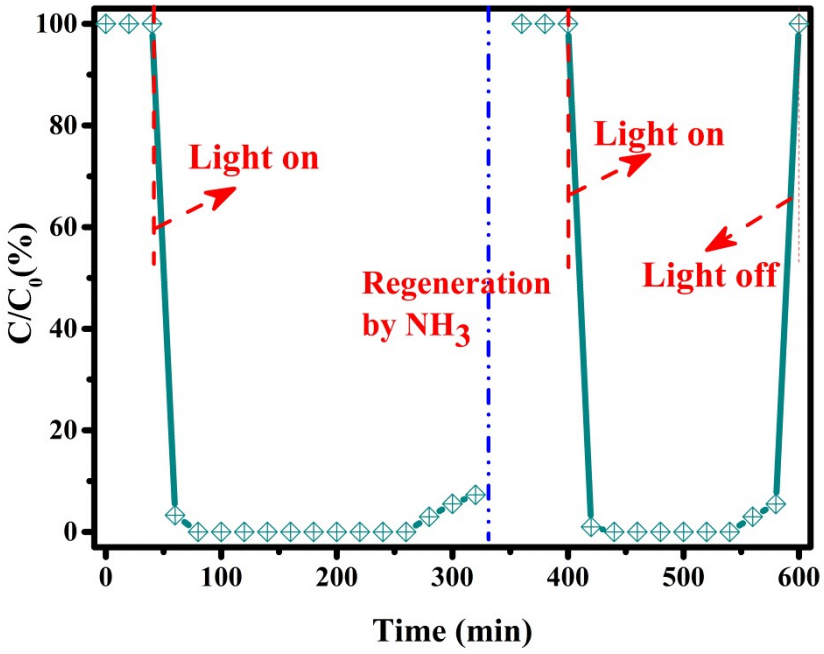


Fig. S3 Long-term stability test for BCN-2 and catalyst regeneration.

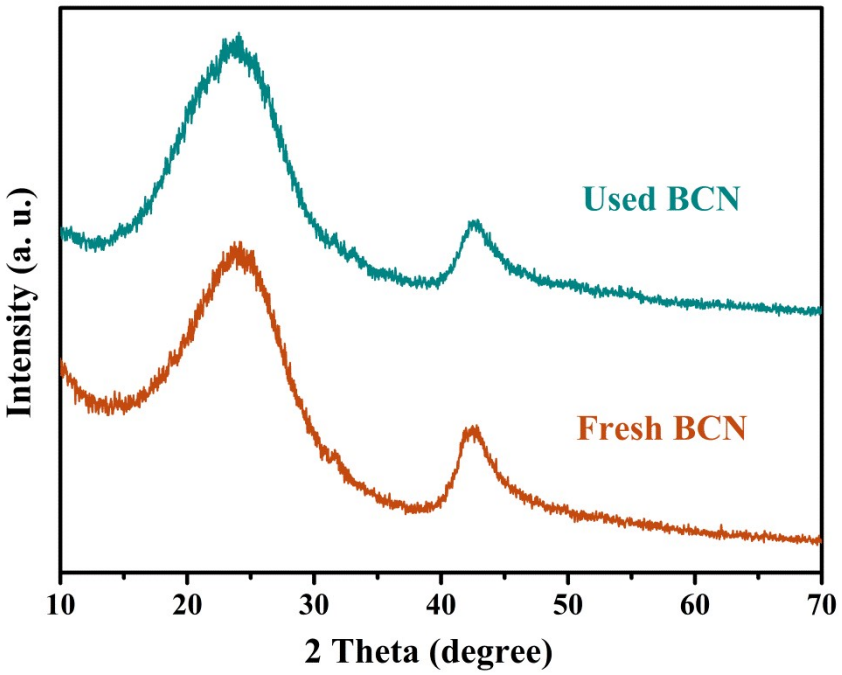


Fig. S4 The XRD patterns of BCN-2 before and after desulfurization.

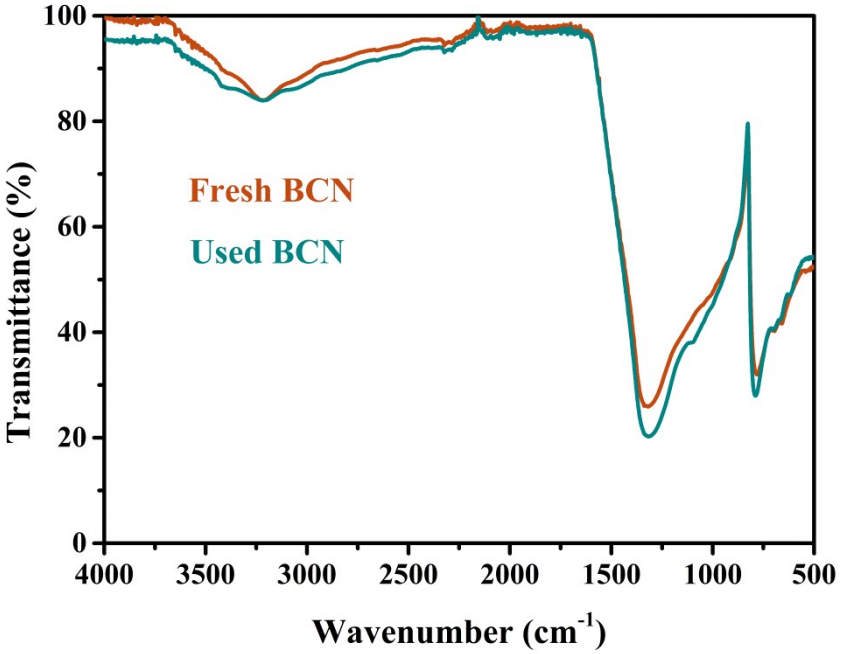


Fig. S5 The FTIR patterns of BCN-2 before and after desulfurization.

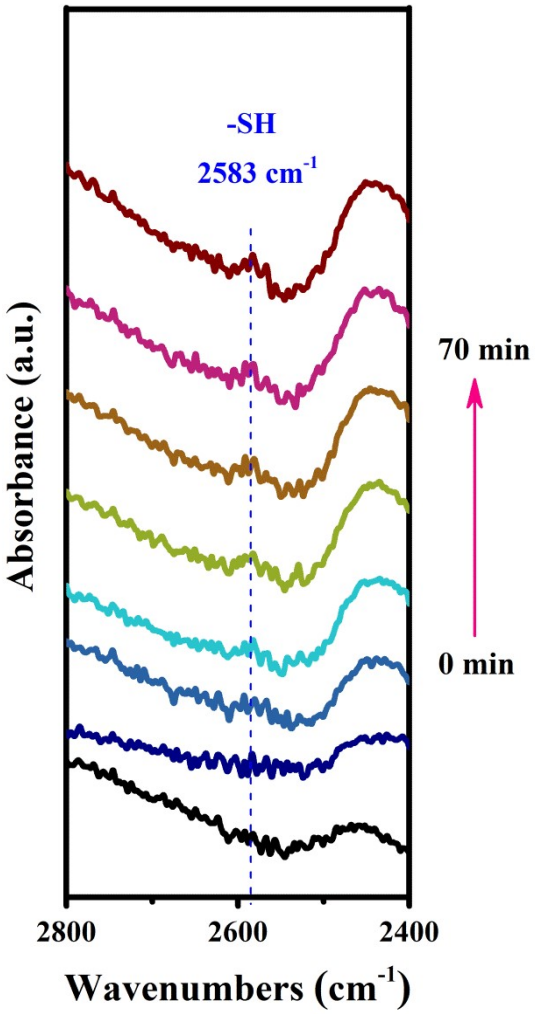


Fig. S6 In situ DRIFTS testing of H_2S adsorption processes over BCN-2 sample.

As the infrared vibrational signal of HS- is very weak, the test was carried out using 50000 ppm of H_2S (N_2 as equilibrium gas) gas. Before testing the sample BCN-2 was heated at 200°C for 2 hours under nitrogen protection to remove surface adsorbed contaminants.

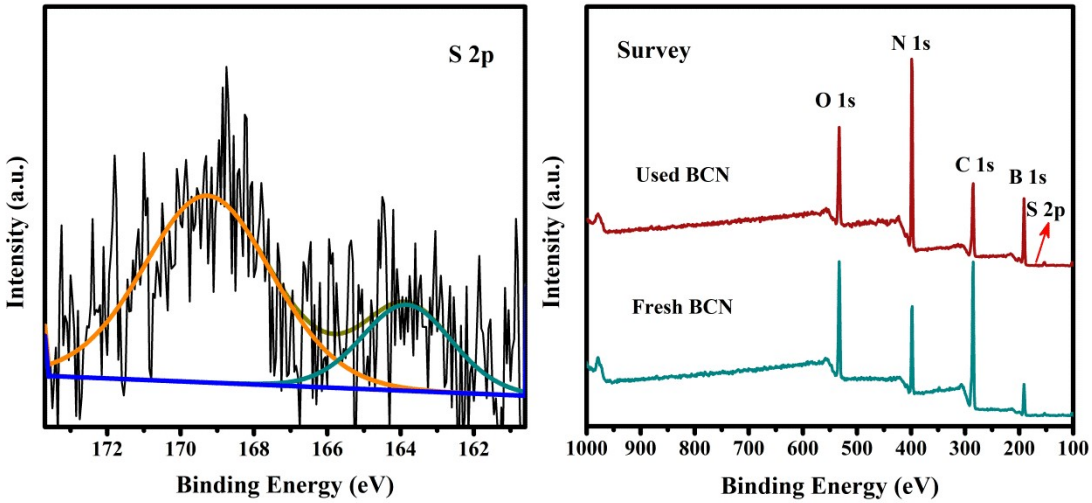


Fig. S7 The residual S on the surface of the BCN-2 is analyzed by XPS after a long-time photocatalytic process.

Table S1 Specific surface area, pore size and atomic percent of the BCN samples (Atomic percentage data from XPS tests).

Samples	S_{BET} (m^2/g)	Pore size (nm)	Atomic %		
			B	N	C
BN	445	2.04	/	/	/
BCN-1	596	2.08	47.38	44.95	7.67
BCN-2	759	2.07	44.55	42.31	13.14
BCN-3	957	2.01	34.13	29.8	36.06

Table S2 Detailed information on the normalization of the activity over BCN-2 under different wavelengths.

	598nm	**530nm**	**495nm**	**470nm**	**420nm**	**380nm**
***P*（mW/cm²)**	52	84	134	241	142	120
C_0 **(ppm)**	21.4	21.4	21.4	21.4	21.3	21.3
C_A **(ppm)**	20.96	20.1	19.54	9.24	0.16	0.04
***η* (%)**	2.056	6.075	8.692	56.822	99.249	99.812
Normalized *η* (%)	1.142	4.821	4.324	15.718	46.596	55.451

Where ***P*** is the optical power density (mW/cm^2), C_0 is the initial H_2S concentration (ppm), C_A is the transient H_2S concentration (ppm), ***η*** is the removal efficiency of H_2S (%) and calculated as ***formula S1***.

$$\eta = (1-C_A/C_0) \times 100\% \qquad (S1)$$

In this work, the illumination area of the catalyst is a constant value of 1.5 cm^2. To ensure the accuracy and reasonable comparison result, the removal efficiency of every point wavelength is normalized by the optical power density. Namely, ***Normalized η*** is the H_2S conversion rate for 100 mW of light irradiation per unit area and is calculated as ***formula S2***.

$$Normalized\ \eta\ (\%) = 100 \times \eta/(P*1.5) \qquad (S2)$$