

**Nitrogen-doped Pitch-based Activated Carbon Fibers with Multi-dimensional Metal Nanoparticle Distribution for The Effective Removal of NO**

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## Characterization

The morphology of the fiber surface and the dispersion of metal nanoparticles were observed by scanning electron microscope (SEM) by a SU 8010 scanning electron microscope (Hitachi, Japan), and combined with the Energy Dispersive Spectroscopy (EDS) in-situ analysis, the element composition was analyzed. Qualitative EDS results were obtained according to the different X-ray signals emitted by each element while quantitative results were obtained according to the intensities of the X-ray signals.

Thermogravimetry (TG) and differential scanning calorimetry (DSC) curves were conducted by a STA 449 F5 thermal analyzer (Netzsch, Germany) at a heating rate of 10 °C/min from room temperature to 1000°C in air to evaluate the approximate content of metal nanoparticles loaded on the fiber surface and to analyze the characteristics of the oxidation process.

X-ray diffraction (XRD) was carried out by a D-5000 automatic X-ray diffractometer (Siemens, Germany) to analyze the phase composition and content of the samples.

X-ray photoelectron spectroscopy (XPS) was performed by a ESCALAB 250Xi X-ray photoelectron spectrometer to compare the chemical changes on the fiber surface. At the same time, the atomic content of Cu2p and Ce3d, and the ratio of  $Ce^{3+}/(Ce^{3+}+Ce^{4+})$  were collected and calculated to analyze the composition and the valence states of metal nanoparticles.

N<sub>2</sub> adsorption-desorption isotherms and pore size distribution curves were measured by a specific surface area tester (ASAP 2460, America). Brunauer-Emmett-Teller (BET) equation was used to calculate the BET specific surface area ( $S_{BET}$ ). *t*-plot method was used to calculate the micropore volume ( $V_{mic}$ ).

NO adsorption properties were measured as follows: 0.05 g N-CNF/ACF@Me was heated to 500°C at 20°C/min and then were cleaned at a constant temperature in N<sub>2</sub>

flow for 1 h. After the samples were cooled down to room temperature, 100 ppm NO, N<sub>2</sub> and 10% O<sub>2</sub> were introduced into the tube at a flow rate of 100 mL/min. The concentration of NO at the outlet was monitored by an analyzer. Meanwhile, the amount of NO adsorbed by each sample within 1 h was roughly calculated according to the following equation:

$$A = \frac{C_0 t_s - \int_0^{t_s} C(t) dt}{C_0 t_s} \times 100\% \approx \left(1 - \frac{S_{curve}}{100 \times 60}\right) \times 100\%$$

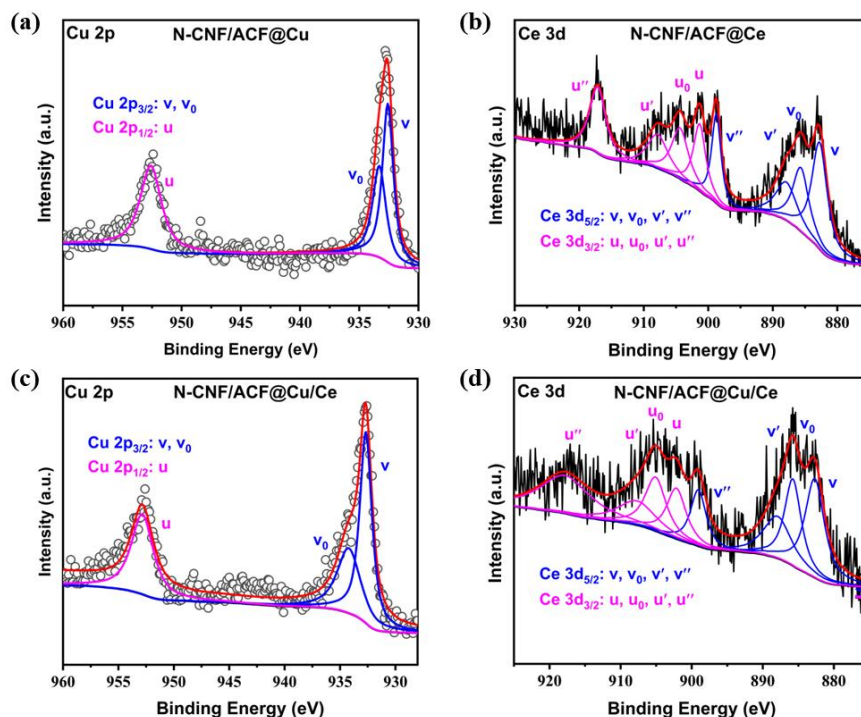
where A is NO adsorption, C<sub>0</sub> and C(t) are the import and export concentrations of NO (ppm), t<sub>s</sub> is time of the adsorbent (min), and S<sub>curve</sub> is the area enclosed by the NO adsorption curve and t axis of 1 h.

The NH<sub>3</sub>-SCR reactivity was evaluated as follows: 500 ppm NO, 500 ppm NH<sub>3</sub> and 5% O<sub>2</sub> were mixed by using N<sub>2</sub> as the equilibrium gas and then passed into the reaction device containing 0.05 g N-CNF/ACF@Me at a total flow rate of 500 mL/min, and the concentration of NO<sub>x</sub> at the outlet was detected by an analyzer.

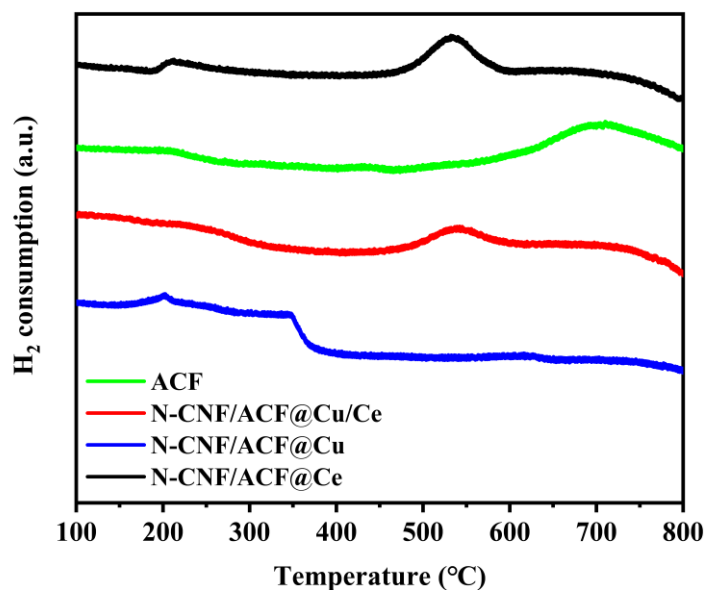
NH<sub>3</sub>-TPD tests was conducted as follows: first, load about 0.04 g sample into the quartz reactor and purg the sample e at room temperature by N<sub>2</sub> at a flow rate of 30 mL/min for 10 min; second, heat the reactor to 200°C at the rate of 10°C/min, holding for 30 min to dewater, and then wait it to cool down to room temperature; third, open the NH<sub>3</sub> gas valve and the rotameter, set the flow rate to 40 mL/min, holding at 80°C for 30 min, then remove the physically adsorbed gas by purging with N<sub>2</sub> at 80 °C for 10 min, and wait for cooling to room temperature; finally (the temperature-programmed desorption step), open the thermal conductivity detector (TCD), and after zero setting and baseline levelling, heat the sample to 1000°C at a heating rate of 10°C/min while using TCD to record the NH<sub>3</sub> desorption synchronously.

H<sub>2</sub>-TPR used the mixture of Ar and H<sub>2</sub> as the carrier gas and N<sub>2</sub> as the washing gas. After removing the residual gas in the sample and dehydrating by heating (the same

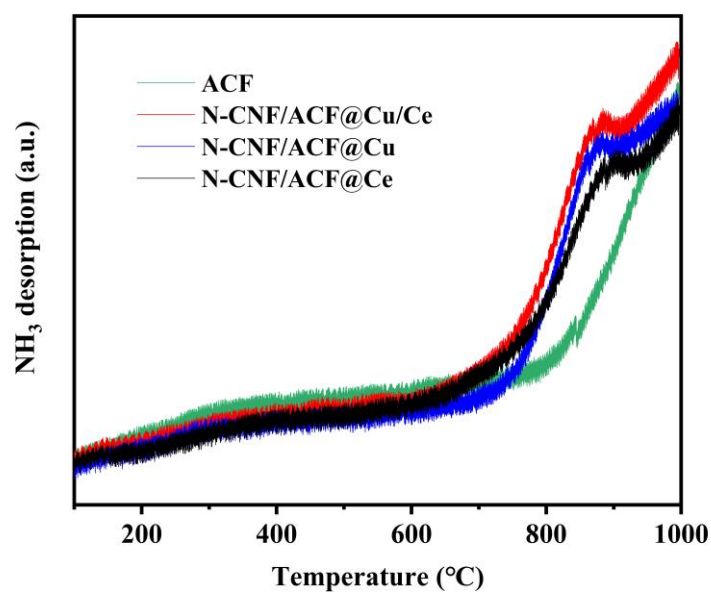
process as  $\text{NH}_3\text{-TPD}$ ), the temperature-programmed reduction step was directly performed, using TCD to record the  $\text{H}_2$  consumption synchronously.



**Figure S1.** XPS peak deconvolution in the  $\text{Cu}2p$  and/or  $\text{Ce}3d$  domains of (a)  $\text{N-CNF/ACF@Cu}$ , (b)  $\text{N-CNF/ACF@Ce}$  and (c, d)  $\text{N-CNF/ACF@Cu/Ce}$ .



**Figure S2.**  $\text{H}_2$ -TPR curves of ACF and  $\text{N-CNF/ACF@Me}$ .



**Figure S3.** NH<sub>3</sub>-TPD curves of ACF and N-CNF/ACF@Me.

**Table S1.** Comparison on NH<sub>3</sub>-SCR reactivity of different materials.

Catalysts	DeNO conditions	NO conversion (%)	Temperature (°C)	Ref.
N-CNF/ACF@Cu/Ce	[NO]=[NH <sub>3</sub> ]=500 ppm [O <sub>2</sub> ]=5% 0.5 g	72~81	295~495	This work
V <sub>2</sub> O <sub>5</sub> -WO <sub>3</sub> -TiO <sub>2</sub>	[NO]=[NH <sub>3</sub> ]=500 ppm [O <sub>2</sub> ]=4% 0.2 g	80~97	300~400	[25]
CuO-CeO <sub>2</sub> -TiO <sub>2</sub>	[NO]=[NH <sub>3</sub> ]=500 ppm [O <sub>2</sub> ]=5% 0.12 g	>95	200~300	[26]
CeO <sub>2</sub> -CuO	[NO]=[NH <sub>3</sub> ]=600 ppm [O <sub>2</sub> ]=5% 1.0 g	>90	160~200	[27]
0.3% CeO <sub>2</sub> /HNO <sub>3</sub> -treated CNTs (0.3Ce-TCNTs)	[NO]=[NH <sub>3</sub> ]=600 ppm [O <sub>2</sub> ]=3.5% 0.5 g	80~95	275~350	[28]
Cu <sub>3.8</sub> -SSZ-13	[NO]=[NH <sub>3</sub> ]=500 ppm [O <sub>2</sub> ]=5%	>95	225~400	[29]